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(54)【発明の名称】　有機エレクトロルミネッセンス素子用材料およびそれを使用した有機エレクトロルミネッセンス素子

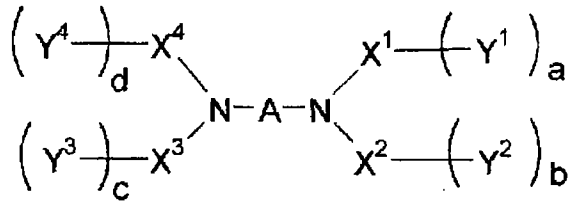
(57)【要約】

【課題】　高輝度で高効率の発光が可能であり、発光劣化が少なく信頼性の高い有機エレクトロルミネッセンス素子用材料およびそれを使用した有機エレクトロルミネッセンス素子を提供する。

【解決手段】下記一般式［１］で示される有機エレクトロルミネッセンス素子用材料。

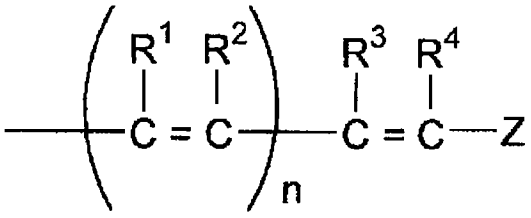
一般式［１］

【化１】



［式中、ＡおよびＸ¹ ～Ｘ⁴ は、それぞれ独立に、置換もしくは未置換の炭素原子数６～２０のアリーレン基を表す。Ｙ¹ ～Ｙ⁴ は、それぞれ独立に、下記一般式［２］で示される有機基を表す。

【化２】



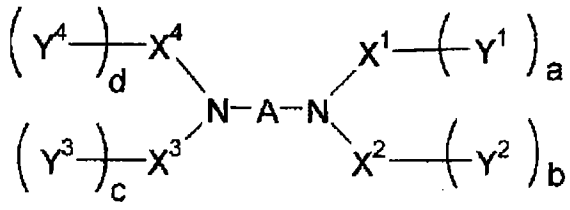
（式中、Ｒ¹ ～Ｒ⁴ は、水素原子、置換もしくは未置換のアルキル基、置換もしくは未置換のアリール基、シアノ基を表すか、Ｒ¹ とＲ² またはＲ³ とＲ⁴ が結合した三重結合を表す。Ｚは、置換もしくは未置換のアリール基を表す。ａ～ｄは、０～２の正の整数を表す。ｎは、０もしくは１を表す。）］

【特許請求の範囲】

【請求項１】　下記一般式［１］で示される有機エレクトロルミネッセンス素子用材料。

一般式［１］

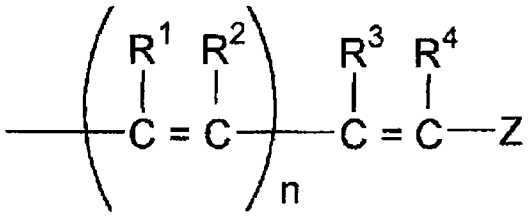
【化１】



［式中、ＡおよびＸ^{1} ～Ｘ^{4} は、それぞれ独立に、置換もしくは未置換の炭素原子数６～２０のアリーレン基を表す。Ｙ^{1} ～Ｙ^{4} は、それぞれ独立に、下記一般式［２］で示される有機基を表す。

一般式［２］

【化２】



（式中、Ｒ^{1} ～Ｒ^{4} は、水素原子、置換もしくは未置換のアルキル基、置換もしくは未置換のアリール基、シアノ基を表すか、Ｒ^{1} とＲ^{2} またはＲ^{3} とＲ^{4} が結合した三重結合を表す。Ｚは、置換もしくは未置換のアリール基を表す。ａ～ｄは、０～２の正の整数を表す。ｎは、０もしくは１を表す。）］

【請求項２】　上記一般式［１］においてＡが、置換もしくは未置換の炭素原子数１０～２０の縮合アリーレン基である請求項１記載の有機エレクトロルミネッセンス素子用材料。

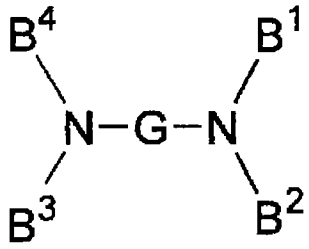
【請求項３】　有機エレクトロルミネッセンス素子用発光材料である請求項１または２記載の有機エレクトロルミネッセンス素子用材料。

【請求項４】　一対の電極間に発光層または発光層を含む複数層の有機化合物薄膜を形成してなる有機エレクトロルミネッセンス素子において、少なくとも一層が請求項１ないし３記載の有機エレクトロルミネッセンス素子用材料を含有する層である有機エレクトロルミネッセンス素子。

【請求項５】　芳香族三級アミン誘導体および／またはフタロシアニン誘導体を含有する層を、発光層と陽極との間に形成してなる請求項４記載の有機エレクトロルミネッセンス素子。

【請求項６】　芳香族三級アミン誘導体が、下記一般式［３］で示される化合物である請求項５記載の有機エレクトロルミネッセンス素子。

【化３】



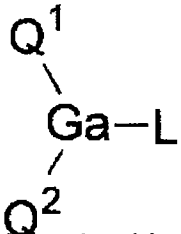
［式中、Ｂ^{1} ～Ｂ^{4} は、それぞれ独立に、置換もしくは未置換の炭素原子数６～２０のアリール基を表す。Ｇは、置換もしくは未置換のアリーレン基を表す。］

【請求項７】　金属錯体化合物もしくは含窒素五員環誘導体を含有する層を、発光層と陰極との間に形成してなる請求項４ないし６記載の有機エレクトロルミネッセンス素子。

【請求項８】　金属錯体化合物が、下記一般式［４］で示される化合物である請求項４ないし７記載の有機エレクトロルミネッセンス素子。

一般式［４］

【化４】



［式中、Ｑ^{1} およびＱ^{2} は、それぞれ独立に、置換もしくは未置換のヒドロキシキノリン誘導体、置換もしくは未置換のヒドロキシベンゾキノリン誘導体を表し、Ｌは、ハロゲン原子、置換もしくは未置換のアルキル基、置換もしくは未置換のシクロアルキル基、置換もしくは未置換の窒素原子を含んでも良いアリール基、－ＯＲ（Ｒは水素原子、置換もしくは未置換のアルキル基、置換もしくは未置換のシクロアルキル基、置換もしくは未置換の窒素原子を含んでも良いアリール基である。）、－Ｏ－Ｇａ－Ｑ^{3} （Ｑ^{4} ）（Ｑ^{3} およびＱ^{4} は、Ｑ^{1} およびＱ^{2}と同じ意味を表す。）で表される配位子を表す。］

【発明の詳細な説明】

【０００１】

【発明の技術分野】本発明は平面光源や表示に使用される有機エレクトロルミネッセンス（ＥＬ）素子用発光材料および高輝度の発光素子に関するものである。

【０００２】

【従来の技術】有機物質を使用したＥＬ素子は、固体発光型の安価な大面積フルカラー表示素子としての用途が有望視され、多くの開発が行われている。一般にＥＬは、発光層および該層をはさんだ一対の対向電極から構成されている。発光は、両電極間に電界が印加される

される。さらに、この電子が発光層において正孔と再結合し、エネルギー準位が伝導帯から価電子帯に戻る際にエネルギーを光として放出する現象である。

【０００３】従来の有機ＥＬ素子は、無機ＥＬ素子に比べて駆動電圧が高く、発光輝度や発光効率も低かった。また、特性劣化も著しく実用化には至っていなかった。近年、１０Ｖ以下の低電圧で発光する高い蛍光量子効率を持った有機化合物を含有した薄膜を積層した有機ＥＬ素子が報告され、関心を集めている（アプライド・フィジクス・レターズ、５１巻、９１３ページ、１９８７年参照）。この方法は、金属キレート錯体を発光層、アミン系化合物を正孔注入層に使用して、高輝度の緑色発光を得ており、６～７Ｖの直流電圧で輝度は数１０００ｃｄ／m^2、最大発光効率は１．５１ｍ／Ｗを達成して、実用領域に近い性能を持っている。

【０００４】しかしながら、現在までの有機ＥＬ素子は、構成の改善により発光強度は改良されているが、未だ充分な発光輝度は有していない。また、繰り返し使用時の安定性に劣るという大きな問題を持っている。これは、例えば、トリス（８－ヒドロキシキノリナート）アルミニウム錯体等の金属キレート錯体が、電界発光時に化学的に不安定であり、陰極との密着性も悪く、短時間の発光で大きく劣化していた。以上の理由により、高い発光輝度、発光効率を持ち、繰り返し使用時での安定性の優れた有機ＥＬ素子の開発のために、優れた発光能力を有し、耐久性のある発光材料の開発が望まれている。

【０００５】

【発明が解決しようとする課題】本発明は、発光輝度が高く、繰り返し使用時での安定性の優れた有機ＥＬ素子の提供にある。本発明者らが鋭意検討した結果、一般式［１］で示される有機ＥＬ素子用材料を少なくとも一層に使用した有機ＥＬ素子の発光輝度および発光効率が高く、繰り返し使用時での安定性も優れていることを見いだし本発明を成すに至った。

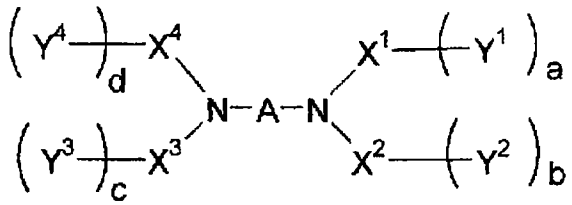
【０００６】

【課題を解決するための手段】本発明は、下記一般式［１］で示される有機エレクトロルミネッセンス素子用材料に関する。

一般式［１］

【０００７】

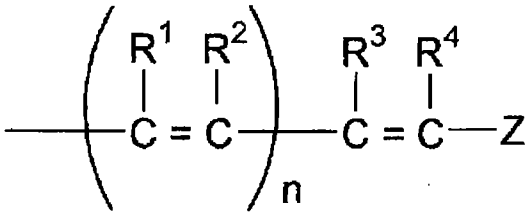
【化５】



【０００８】［式中、ＡおよびX¹ ～X⁴ は、それぞれ独立に、置換もしくは未置換の炭素原子数６～２０のアリーレン基を表す。Ｙ¹ ～Ｙ⁴ は、それぞれ独立に、置換もしくは未置換の炭素原子数６～２０のアリール基を表す。ａ～ｄは、０～２の正の整数を表す。］

【０００９】

【化６】



【００１０】（式中、Ｒ¹ ～Ｒ⁴ は、水素原子、置換もしくは未置換のアルキル基、置換もしくは未置換のアリール基、シアノ基を表すか、Ｒ¹ とＲ² またはＲ³ とＲ⁴ が結合した三重結合を表す。Ｚは、置換もしくは未置換のアリール基を表す。ａ～ｄは、０～２の正の整数を表す。ｎは、０もしくは１を表す。）］

【００１１】さらに本発明は、上記一般式［１］においてＡが、置換もしくは未置換の炭素原子数１０～２０の縮合アリーレン基である上記有機エレクトロルミネッセンス素子用材料である。

【００１２】さらに本発明は、上記一般式［１］で示される有機エレクトロルミネッセンス素子用発光材料である。

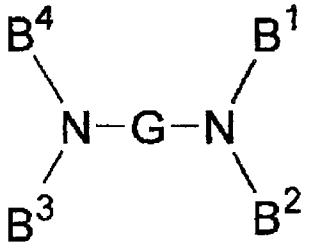
【００１３】さらに本発明は、一対の電極間に発光層または発光層を含む複数層の有機化合物薄膜を形成してなる有機エレクトロルミネッセンス素子において、少なくとも一層が上記有機エレクトロルミネッセンス素子用材料を含有する層である有機エレクトロルミネッセンス素子である。

【００１４】さらに本発明は、芳香族三級アミン誘導体および／またはフタロシアニン誘導体を含有する層を、発光層と陽極との間に形成してなる上記有機エレクトロルミネッセンス素子である。

【００１５】さらに本発明は、芳香族三級アミン誘導体が、下記一般式［３］で示される化合物である上記有機エレクトロルミネッセンス素子である。

一般式［３］

【化７】



［式中、Ｂ¹ ～Ｂ⁴ は、それぞれ独立に、置換もしくは未置換の炭素原子数６～２０のアリール基を表す。Ｇは、置換もしくは未置換のアリーレン基を表す。］

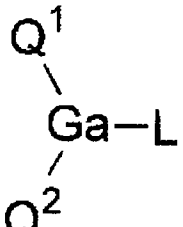
【００１６】さらに本発明は、金属錯体化合物もしくは

間に形成してなる上記有機エレクトロルミネッセンス素子である。

【００１７】さらに本発明は、金属錯体化合物が、下記一般式［４］で示される化合物である上記有機エレクトロルミネッセンス素子である。

一般式［４］

【化８】



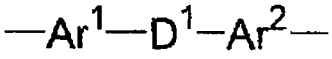
［式中、Q^1 および Q^2 は、それぞれ独立に、置換もしくは未置換のヒドロキシキノリン誘導体、置換もしくは未置換のヒドロキシベンゾキノリン誘導体を表し、Ｌは、ハロゲン原子、置換もしくは未置換のアルキル基、置換もしくは未置換のシクロアルキル基、置換もしくは未置換の窒素原子を含んでも良いアリール基、－ＯＲ（Ｒは水素原子、置換もしくは未置換のアルキル基、置換もしくは未置換のシクロアルキル基、置換もしくは未置換の窒素原子を含んでも良いアリール基である。）、－Ｏ－Ｇａ－Q^3（Q^4）（Q^3 および Q^4 は、Q^1 および Q^2 と同じ意味を表す。）で表される配位子を表す。］

【発明の実施の形態】

【００１８】本発明における一般式［１］で示される化合物のＡおよびX^1～X^4は、それぞれ独立に置換もしくは未置換の炭素原子数６～２０のアリーレン基を表す。ＡおよびX^1～X^4の具体例は、フェニレン基、ビフェニレン基、ターフェニレン基、ナフチレン基、アントリレン基、フェナントリレン基、フルオレニレン基、ピレニレン基、チオフェニレン基等のアリーレン基および下記一般式［５］もしくは一般式［６］で示されるアリーレン基であるがこれらに限定されるものではない。また、炭素原子数１０～２０の縮合アリーレン基は、ナフチレン基、アントリレン基、フェナントリレン基、フルオレニレン基、ピレニレン基のように炭素原子数１０～２０からなる二価の縮合芳香族環を表す。

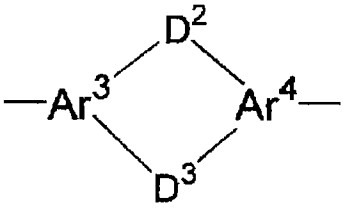
【００１９】一般式［５］

【化９】



【００２０】一般式［６］

【化１０】



【００２１】ここで、一般式［５］および一般式［６］のAr^1～Ar^4は、それぞれ独立に置換もしくは未置換の炭素原子数６～２０のアリーレン基を表す。Ar^1～Ar^4の具体例は、フェニレン基、ビフェニレン基、ターフェニレン基、ナフチレン基、アントリレン基、フェナントリレン基、フルオレニレン基、ピレニレン基、チオフェニレン基等のアリーレン基である。D^1～D^3は、直接結合または、Ｃ、Ｎ、Ｈ、Ｏ、Ｓから選ばれる１～５０個の原子で構成される化学的に合理的な組合わせからなる二価の結合基である。

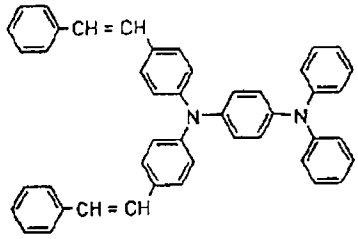
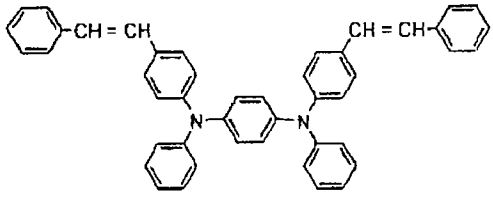
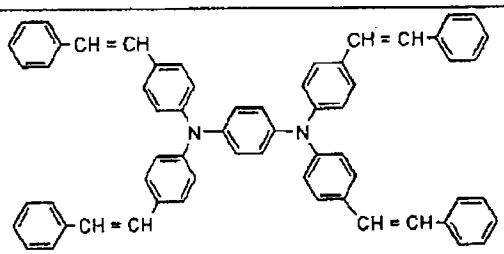
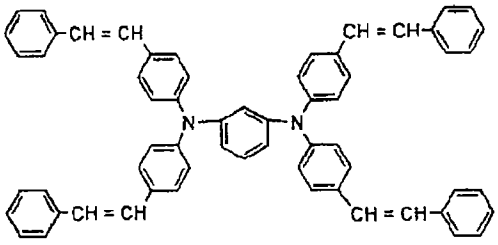
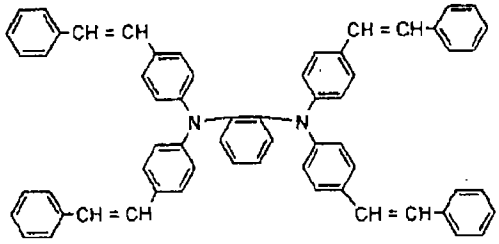
【００２２】本発明における一般式［２］で示される化合物のR^1～R^4は、それぞれ独立に、水素原子、置換もしくは未置換のアルキル基、置換もしくは未置換のアリール基もしくはシアノ基を表す。R^1～R^4の具体例は、置換もしくは未置換のアルキル基としては、メチル基、エチル基、プロピル基、ブチル基、ｓｅｃ－ブチル基、ｔｅｒｔ－ブチル基、ペンチル基、ヘキシル基、ヘプチル基、オクチル基、ステアリル基、２－フェニルイソプロピル基、トリクロロメチル基、トリフルオロメチル基、ベンジル基、α－フェノキシベンジル基、α，α－ジメチルベンジル基、α，α－メチルフェニルベンジル基、α，α－ジトリフルオロメチルベンジル基、トリフェニルメチル基、α－ベンジルオキシベンジル基等がある。置換もしくは未置換のアリール基としては、フェニル基、２－メチルフェニル基、３－メチルフェニル基、４－メチルフェニル基、４－エチルフェニル基、ビフェニル基、４－メチルビフェニル基、４－エチルビフェニル基、４－シクロヘキシルビフェニル基ターフェニル基、３，５－ジクロロフェニル基、ナフチル基、５－メチルナフチル基、アントリル基、ピレニル基等がある。一般式［１］において、ａ～ｄはそれぞれ独立に０～２の正の整数を表し、ｎはそれぞれ独立に０もしくは１を表す。

【００２３】本発明における一般式［２］で示される化合物のＺは、それぞれ独立に置換もしくは未置換の炭素原子数６～２０のアリール基を表す。Ｚの具体例は、フェニル基、ビフェニル基、ターフェニル基、ナフチル基、アントリル基、フェナントリル基、フルオレニル基、ピレニル基、チオフェン基等のアリール基であり、上記アリール基は置換基を有していても良い。置換基の具体例は、R^1～R^4で記述したアルキル基およびアリール基に加えて、アルコキシ基、アミノ基、シアノ基、水酸基、カルボン酸基、エーテル基、エステル基等があ

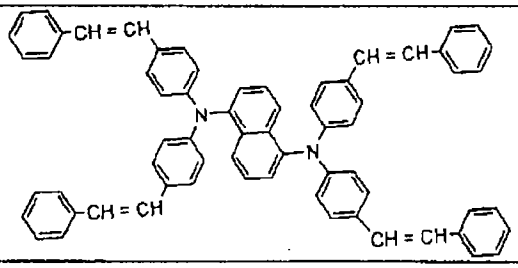
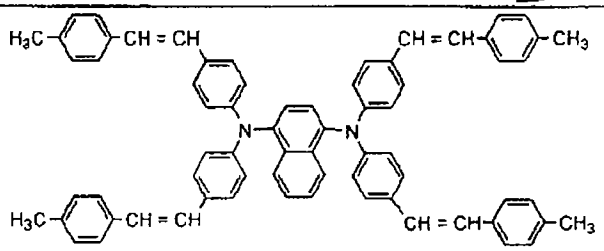
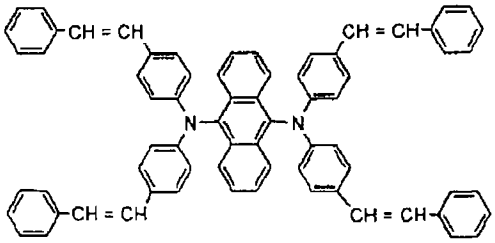
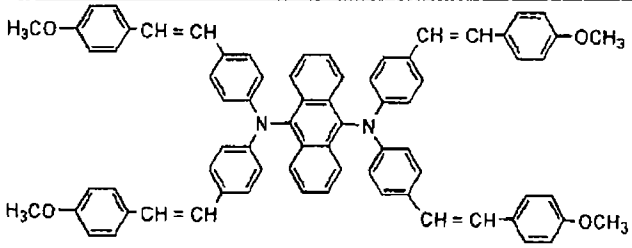
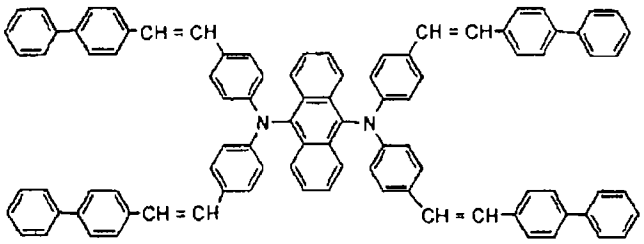
【００２４】以下に、本発明の一般式［１］の化合物の代表例を、表１に具体的に例示するが、本発明は、この代表例に限定されるものではない。

【００２５】

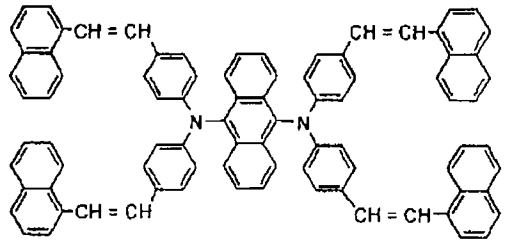
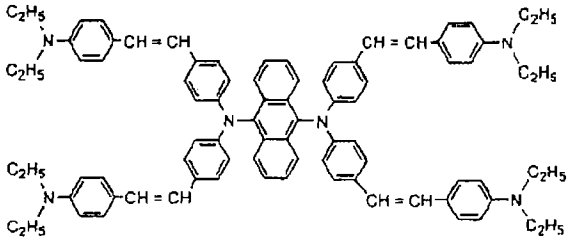
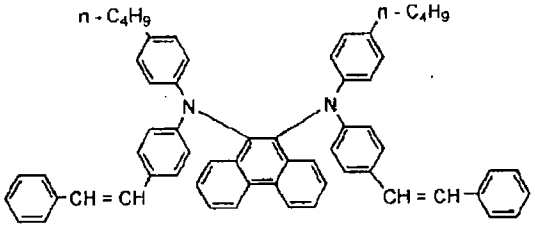
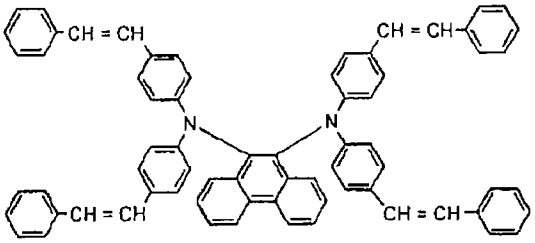
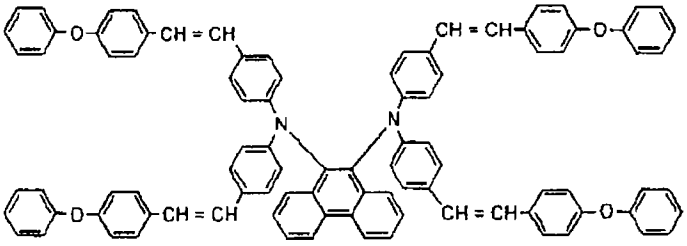
【表１】

化合物	化学構造式
（１）	
（２）	
（３）	
（４）	
（５）	

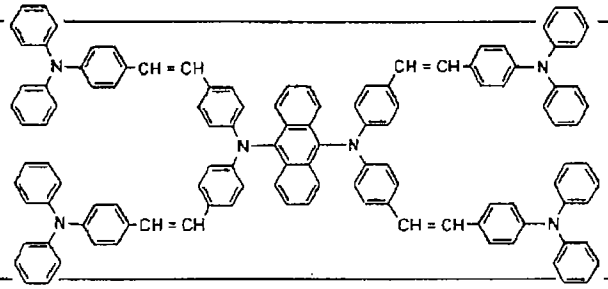
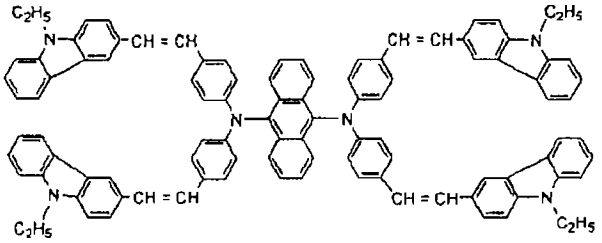
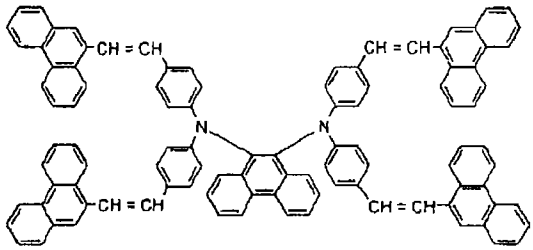
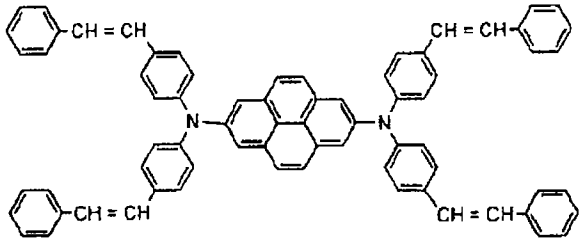
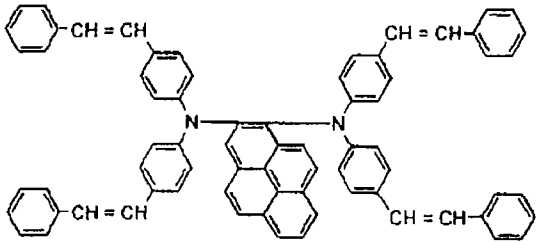
【００２６】

化合物	化 学 構 造 式
(6)	
(7)	
(8)	
(9)	
(10)	

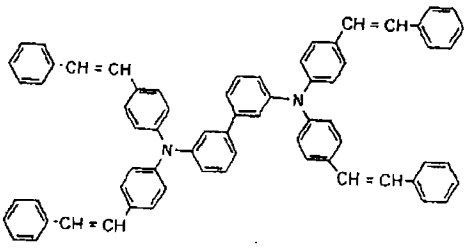
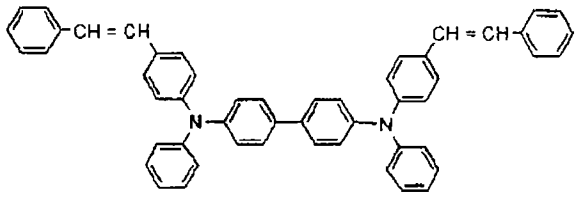
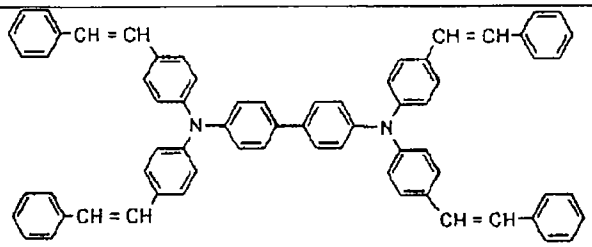
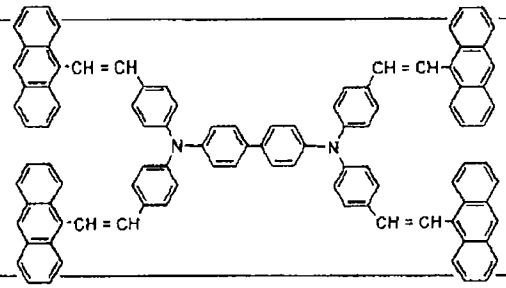
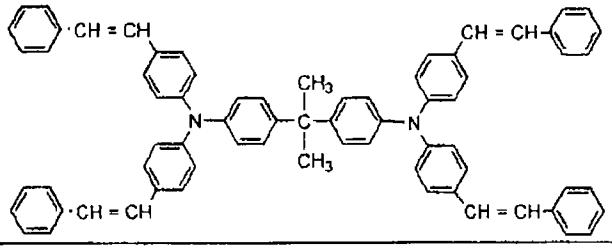
【0027】

化合物	化学構造式
(11)	
(12)	
(13)	
(14)	
(15)	

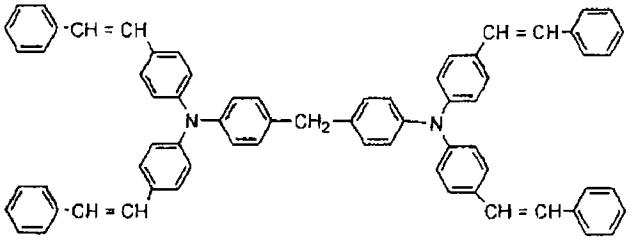
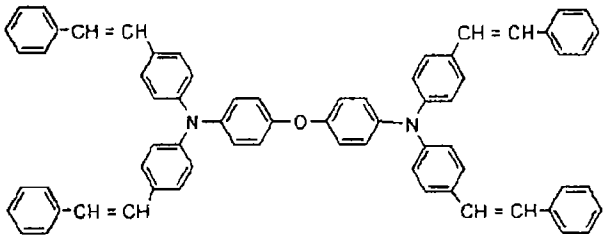
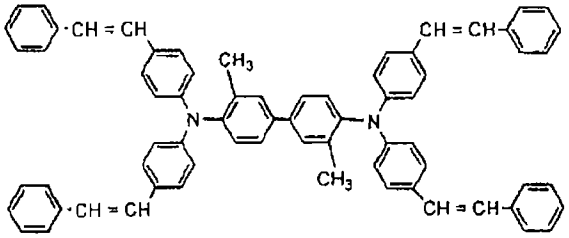
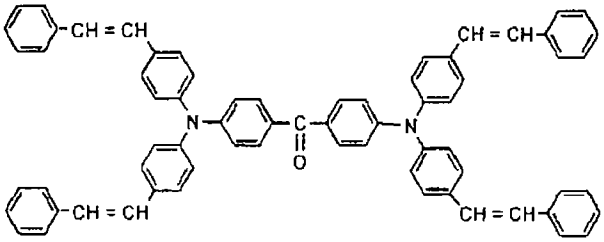
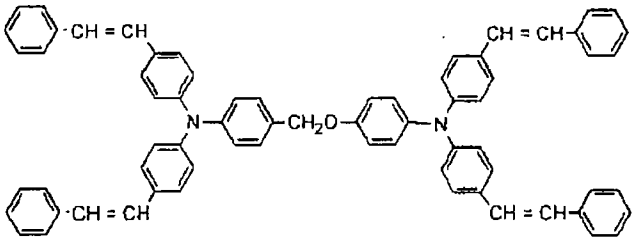
【0028】

化合物	化学構造式
(16)	
(17)	
(18)	
(19)	
(20)	

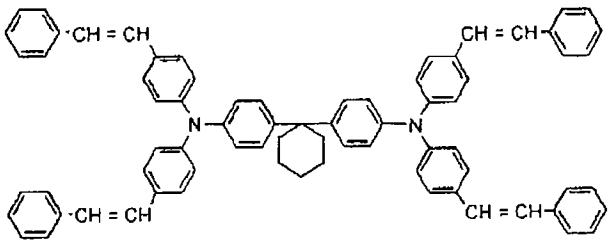
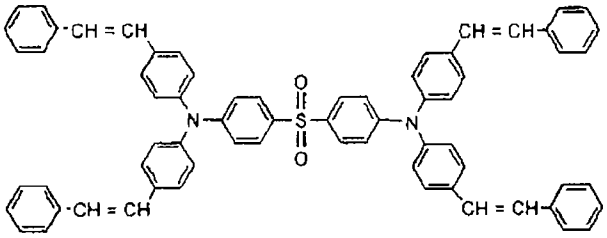
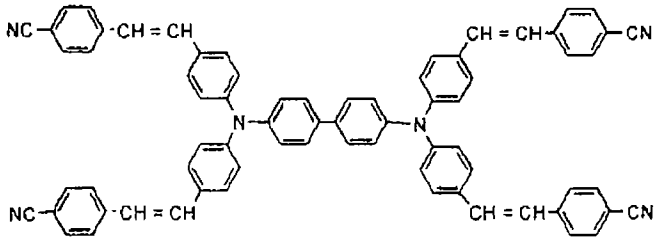
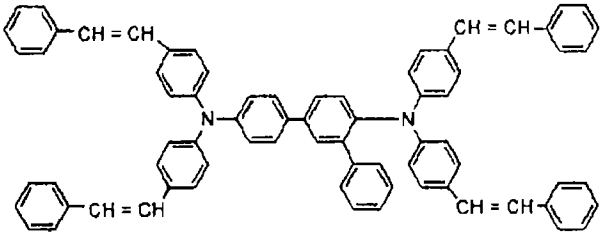
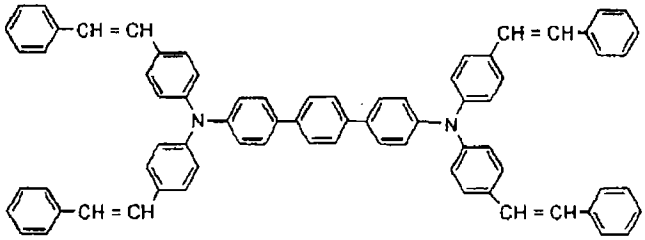
【0029】

化合物	化　学　構　造　式
（２１）	
（２２）	
（２３）	
（２４）	
（２５）	

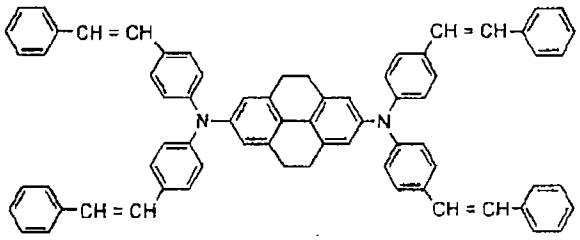
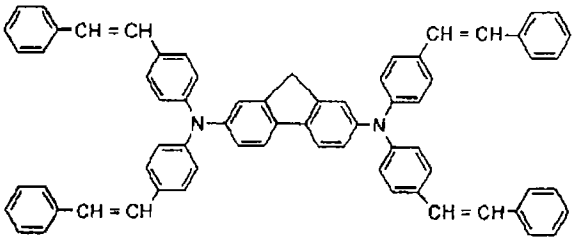
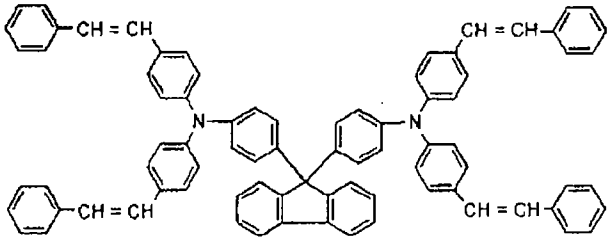
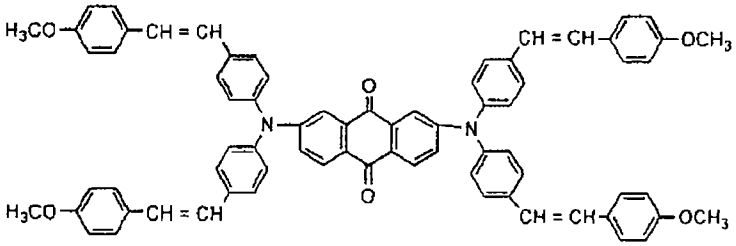
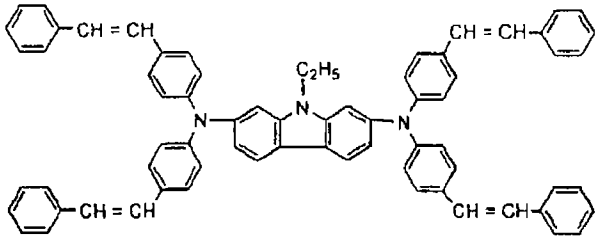
【００３０】

化合物	化学構造式
(26)	
(27)	
(28)	
(29)	
(30)	

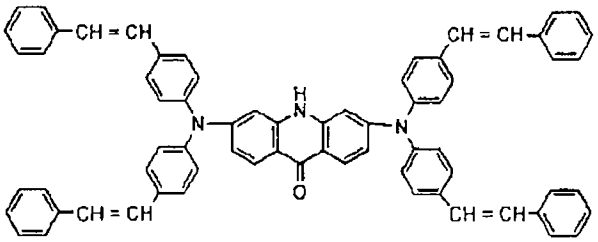
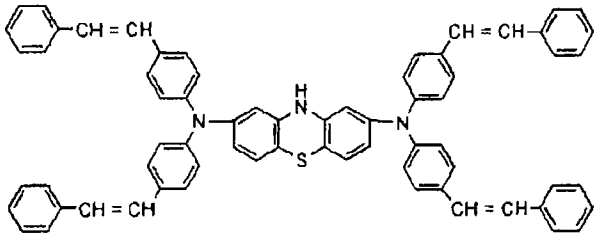
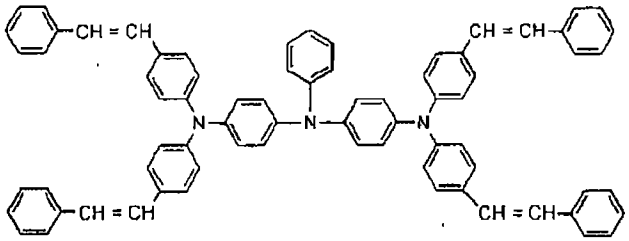
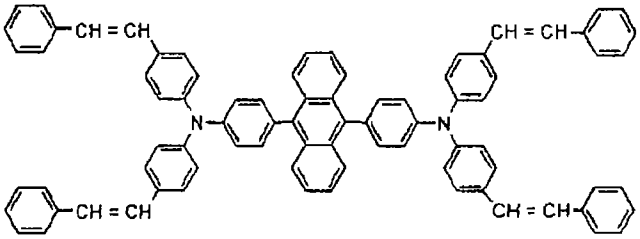
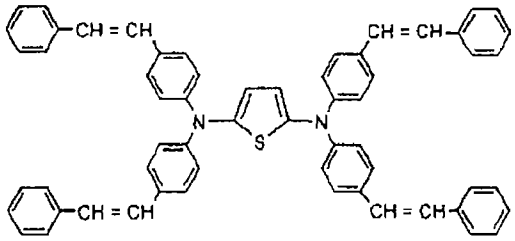
【0031】

化合物	化学構造式
(31)	
(32)	
(33)	
(34)	
(35)	

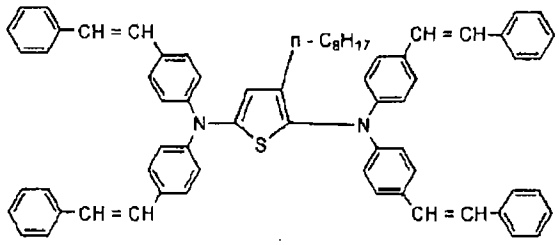
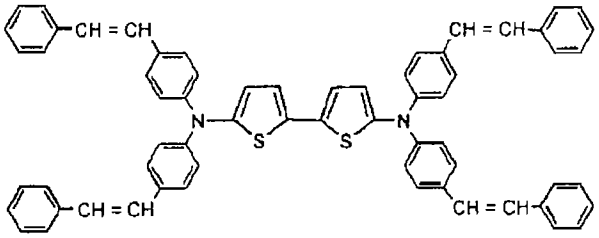
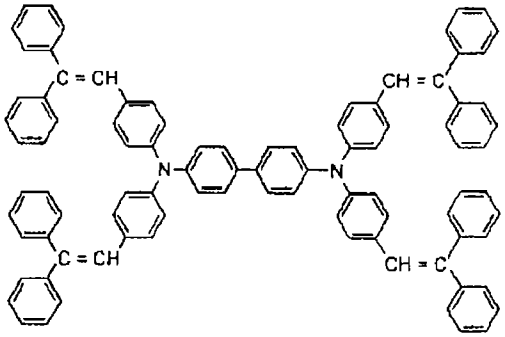
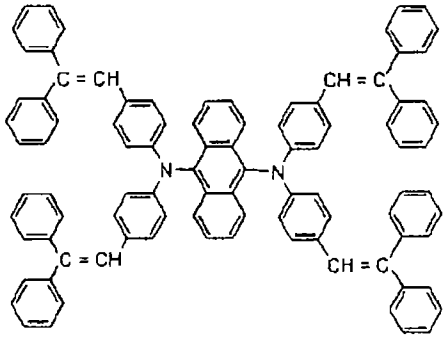
【0032】

化合物	化学構造式
(36)	
(37)	
(38)	
(39)	
(40)	

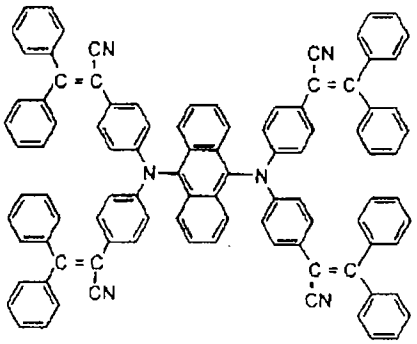
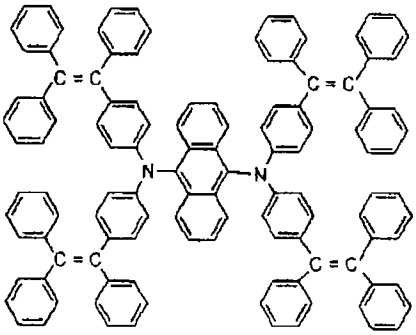
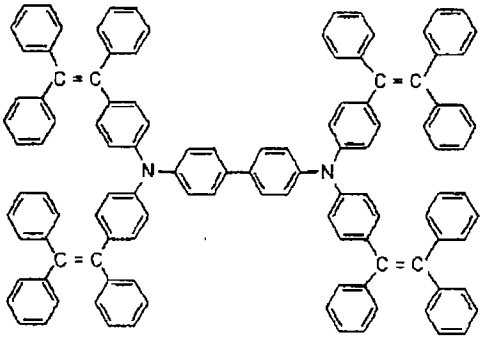
【0033】

化合物	化学構造式
(41)	
(42)	
(43)	
(44)	
(45)	

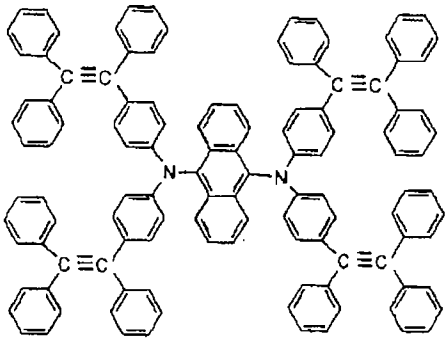
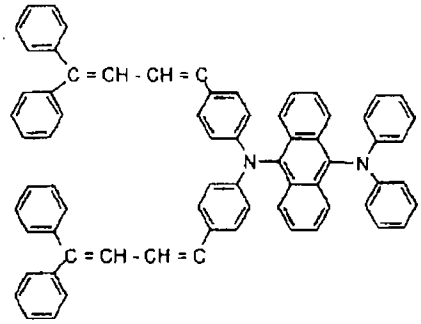
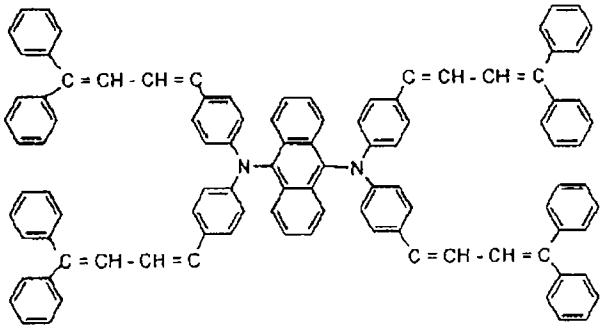
【0034】

化合物	化学構造式
(46)	
(47)	
(48)	
(49)	

【0035】

化合物	化学構造式
(50)	
(51)	
(52)	

【0036】

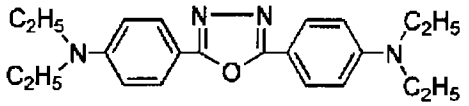
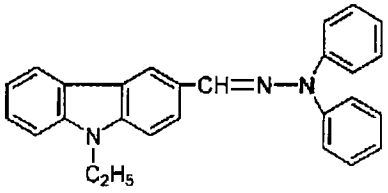
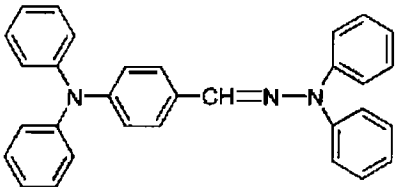
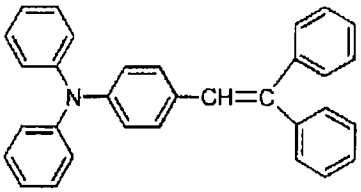
化合物	化学構造式
(53)	
(54)	
(55)	

【００３７】本発明における一般式［３］で示される化合物のB^{1}～B^{4}の具体例は、置換もしくは未置換の炭素原子数６～２０のアリール基である。具体的には、フェニル基、ビフェニル基、ターフェニル基、ナフチル基、アントリル基、フェナントリル基、フルオレニル基、ピレニル基等の窒素原子を含有しても良いアリール基であり、それぞれのアリール基は置換基を有していても良い。Gは、二価のアリーレン基であり、フェニレン基、ビフェニレン基、ターフェニレン基、ナフチレン基、アントリレン基、フェナントリレン基、フルオレニレン基、ピレニレン基等の窒素原子を含有しても良い二価のアリーレン基であり、それぞれのアリール基は置換基を有していても良い。

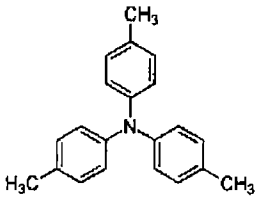
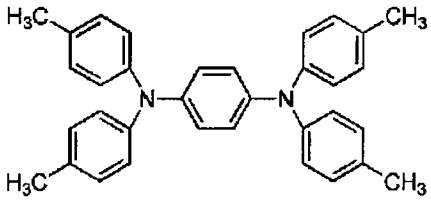
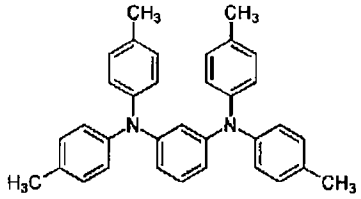
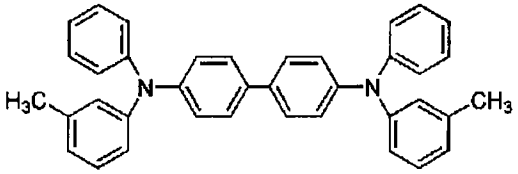
【００３８】以下に、効果的な正孔注入材料である本発明の一般式［３］の化合物およびその他の材料の代表例を、表２に具体的に例示するが、本発明は、この代表例に限定されるものではない。

【００３９】

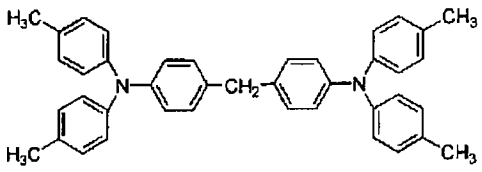
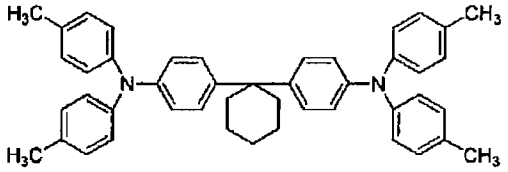
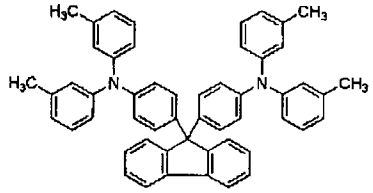
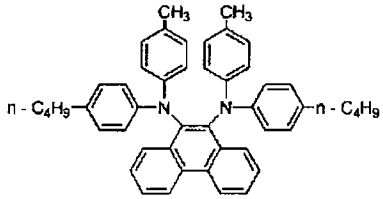
【表２】

化合物	化 学 構 造
A－1	
A－2	
A－3	
A－4	

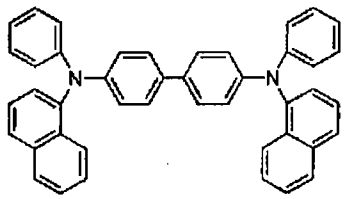
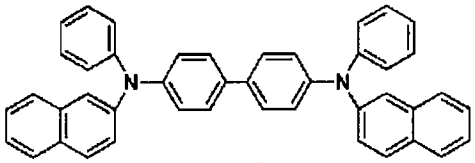
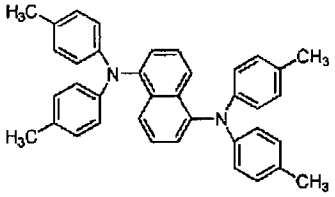
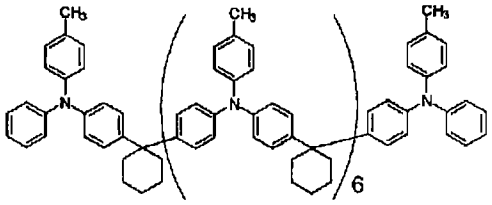
【００４０】

化合物	化 学 構 造
A－5	
A－6	
A－7	
A－8	

【００４１】

化合物	化　学　構　造
A－9	
A－10	
A－11	
A－12	

【0042】

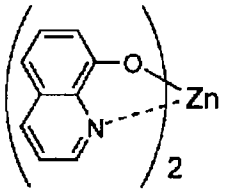
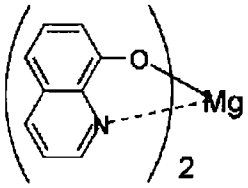
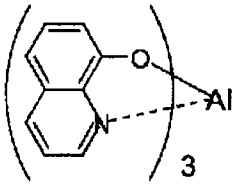
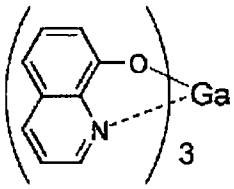
化合物	化学構造
A－13	
A－14	
A－15	
A－16	

【００４３】本発明における一般式［４］で示される化合物のQ^1、Q^{4}は、８－ヒドロキシキノリン、８－ヒドロキシキナルジン、８－ヒドロキシ－２－フェニルキノリン、８－ヒドロキシ－５－メチルキノリン、８－ヒドロキシ－３，５，７－トリフルオロキノリン等のヒドロキシキノリン誘導体、Ｌは、ハロゲン原子、置換もしくは未置換のアルキル基、置換もしくは未置換のシクロアルキル基、置換もしくは未置換の窒素原子を含んでも良いアリール基、－ＯＲ（Ｒは水素原子、置換もしくは未置換のアルキル基、置換もしくは未置換のシクロアルキル基、置換もしくは未置換の窒素原子を含んでも良いアリール基である。）、－Ｏ－Ｇａ－Q^3（Q^4）（Q^{3}およびQ^{4}は、Q^{1}およびQ^{2}と同じ意味を表す。）を示す。ここで、ハロゲン原子、アルキル基、シクロアルキル基、窒素原子を含んでも良いアリール基、および－ＯＲ基のＲのアルキル基、シクロアルキル基、窒素原子を含んでも良いアリール基は、前記の一般式［２］で記述したＲ^1～Ｒ^{4}と同様の基を表す。

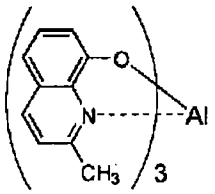
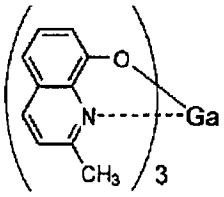
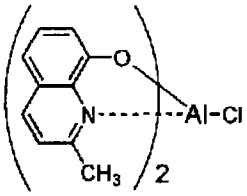
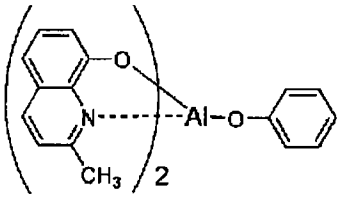
【００４４】以下に、本発明の有機ＥＬ素子に使用する一般式［４］の化合物の代表例および電子注入材料の代表例を、表３に具体的に例示するが、本発明は、この代表例に限定されるものではない。

【００４５】

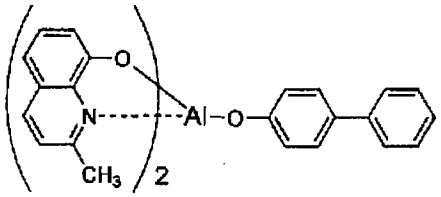
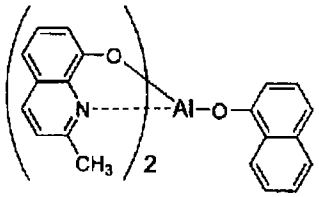
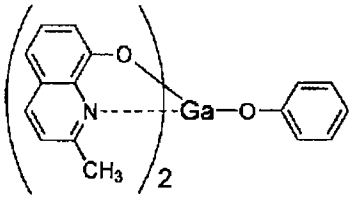
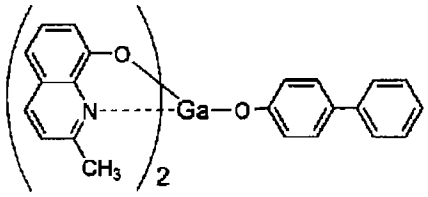
【表３】

化合物	化 学 構 造
B－1	
B－2	
B－3	
B－4	

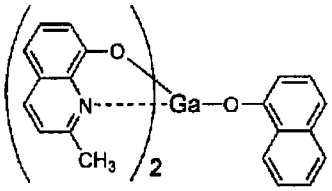
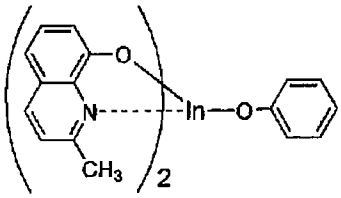
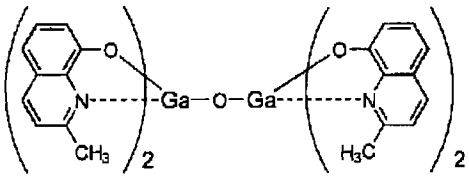
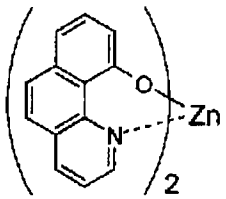
【００４６】

化合物	化 学 構 造
B－5	
B－6	
B－7	
B－8	

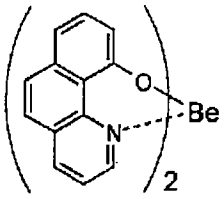
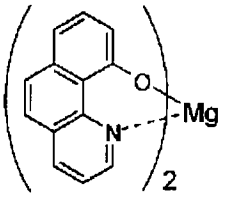
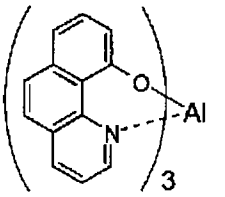
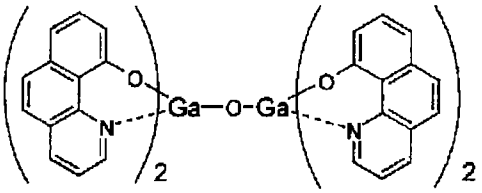
【００４７】

化合物	化学構造
B－9	
B－10	
B－11	
B－12	

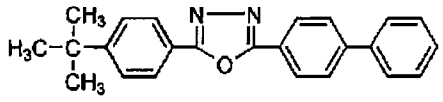
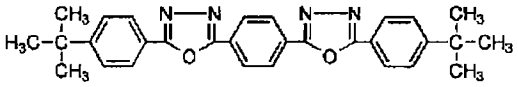
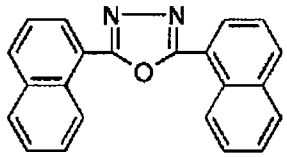
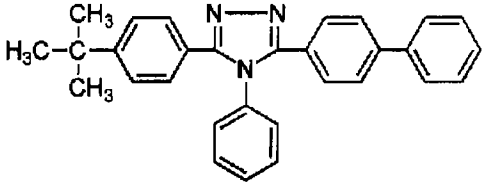
【0048】

化合物	化 学 構 造
B－１３	
B－１４	
B－１５	
B－１６	

【００４９】

化合物	化学構造
B－17	
B－18	
B－19	
B－20	

【0050】

化合物	化学構造
B－２１	
B－２２	
B－２３	
B－２４	

【００５１】本発明の一般式［１］で示される化合物は、固体状態において強い蛍光を持つ化合物であり電場発光性にも優れている。また、金属電極もしくは有機薄膜層からの優れた正孔注入性および正孔輸送性、金属電極もしくは有機薄膜層からの優れた電子注入性および電子輸送性を併せて持ち合わせているので、発光材料として有効に使用することができ、更には、他の正孔輸送性材料、電子輸送性材料もしくはドーピング材料を使用してもさしつかえない。

【００５２】有機ＥＬ素子は、陽極と陰極間に一層もしくは多層の有機薄膜を形成した素子である。一層型の場合、陽極と陰極との間に発光層を設けている。発光層は、発光材料を含有し、それに加えて陽極から注入した正孔、もしくは陰極から注入した電子を発光材料まで輸送させるために、正孔注入材料もしくは電子注入材料を含有しても良い。しかしながら、本発明の発光材料は、極めて高い発光量子効率、高い正孔輸送能力および電子輸送能力を併せ持ち、均一な薄膜を形成することができるので、本発明の発光材料のみで発光層を形成することも可能である。多層型は、（陽極／正孔注入層／発光層／陰極）、（陽極／発光層／電子注入層／陰極）、（陽極／正孔注入層／発光層／電子注入層／陰極）の多層構成で積層した有機ＥＬ素子がある。一般式［１］の化合物は、高い発光特性を持ち、正孔注入性、正孔輸送特性光材料として発光層に使用することができる。

【００５３】発光層には、必要があれば、本発明の一般式［１］の化合物に加えて、さらなる公知の発光材料、ドーピング材料、正孔注入材料や電子注入材料を使用することもできる。有機ＥＬ素子は、多層構造にすることにより、クエンチングによる輝度や寿命の低下を防ぐことができる。必要があれば、発光材料、ドーピング材料、正孔注入材料や電子注入材料を組み合わせて使用することが出来る。また、ドーピング材料により、発光輝度や発光効率の向上、赤色や青色の発光を得ることもできる。また、正孔注入層、発光層、電子注入層は、それぞれ二層以上の層構成により形成されても良い。その際には、正孔注入層の場合、電極から正孔を注入する層を正孔注入層、正孔注入層から正孔を受け取り発光層まで正孔を輸送する層を正孔輸送層と呼ぶ。同様に、電子注入層の場合、電極から電子を注入する層を電子注入層、電子注入層から電子を受け取り発光層まで電子を輸送する層を電子輸送層と呼ぶ。これらの各層は、材料のエネルギー準位、耐熱性、有機層もしくは金属電極との密着性等の各要因により選択されて使用される。

【００５４】一般式［１］の化合物と共に発光層に使用できる発光材料またはドーピング材料としては、アントラセン、ナフタレン、フェナントレン、ピレン、テトラセン、コロネン、クリセン、フルオレセイン、ペリレ

タロペリノン、ナフタロペリノン、ジフェニルブタジエン、テトラフェニルブタジエン、クマリン、オキサジアゾール、アルダジン、ビスベンゾキサゾリン、ビススチリル、ピラジン、シクロペンタジエン、キノリン金属錯体、アミノキノリン金属錯体、ベンゾキノリン金属錯体、イミン、ジフェニルエチレン、ビニルアントラセン、ジアミノカルバゾール、ピラン、チオピラン、ポリメチン、メロシアニン、イミダゾールキレート化オキシノイド化合物、キナクリドン、ルブレンおよび蛍光色素等があるが、これらに限定されるものではない。

【００５５】正孔注入材料としては、正孔を輸送する能力を持ち、陽極からの正孔注入効果、発光層または発光材料に対して優れた正孔注入効果を有し、発光層で生成した励起子の電子注入層または電子注入材料への移動を防止し、かつ薄膜形成能力の優れた化合物が挙げられる。具体的には、フタロシアニン誘導体、ナフタロシアニン誘導体、ポルフィリン誘導体、オキサゾール、オキサジアゾール、トリアゾール、イミダゾール、イミダゾロン、イミダゾールチオン、ピラゾリン、ピラゾロン、テトラヒドロイミダゾール、オキサゾール、オキサジアゾール、ヒドラゾン、アシルヒドラゾン、ポリアリールアルカン、スチルベン、ブタジエン、ベンジジン型トリフェニルアミン、スチリルアミン型トリフェニルアミン、ジアミン型トリフェニルアミン等と、それらの誘導体、およびポリビニルカルバゾール、ポリシラン、導電性高分子等の高分子材料等があるが、これらに限定されるものではない。

【００５６】本発明の有機ＥＬ素子において使用できる正孔注入材料の中で、さらに効果的な正孔注入材料は、一般式［３］で示した芳香族三級アミン誘導体もしくはフタロシアニン誘導体である。具体的には、トリフェニルアミン、トリトリルアミン、トリルジフェニルアミン、Ｎ，Ｎ’－ジフェニル－Ｎ，Ｎ’－（３－メチルフェニル）－１，１’－ビフェニル－４，４’－ジアミン、Ｎ，Ｎ，Ｎ’，Ｎ’－（４－メチルフェニル）－１，１’－フェニル－４，４’－ジアミン、Ｎ，Ｎ，Ｎ’，Ｎ’－（４－メチルフェニル）－１，１’－ビフェニル－４，４’－ジアミン、Ｎ，Ｎ’－ジフェニル－Ｎ，Ｎ’－ジナフチル－１，１’－ビフェニル－４，４’－ジアミン、Ｎ，Ｎ’－（メチルフェニル）－Ｎ，Ｎ’－（４－ｎ－ブチルフェニル）－フェナントレン－９，１０－ジアミン、Ｎ，Ｎ－ビス（４－ジ－４－トリルアミノフェニル）－４－フェニル－シクロヘキサン等、もしくはこれらの芳香族三級アミン骨格を有したオリゴマーもしくはポリマー等があるが、これらに限定されるものではない。

【００５７】フタロシアニン（Ｐｃ）誘導体としては、H_2 Ｐｃ、ＣｕＰｃ、ＣｏＰｃ、ＮｉＰｃ、ＺｎＰｃ、ＰｄＰｃ、ＦｅＰｃ、ＭｎＰｃ、ＣｌＡｌＰｃ、ＣｌＧａＰｃ、（ＨＯ）ＡｌＰｃ、（ＨＯ）ＧａＰｃ、ＶＯＰｃ、ＴｉＯＰｃ、ＭｏＯＰｃ、ＧａＰｃ－Ｏ－ＧａＰｃ等のフタロシアニン誘導体およびナフタロシアニン誘導体等があるが、これらに限定されるものではない。

【００５８】電子注入材料としては、電子を輸送する能力を持ち、陰極からの正孔注入効果、発光層または発光材料に対して優れた電子注入効果を有し、発光層で生成した励起子の正孔注入層への移動を防止し、かつ薄膜形成能力の優れた化合物が挙げられる。例えば、フルオレノン、アントラキノジメタン、ジフェノキノン、チオピランジオキシド、オキサゾール、オキサジアゾール、トリアゾール、イミダゾール、ペリレンテトラカルボン酸、フレオレニリデンメタン、アントラキノジメタン、アントロン等とそれらの誘導体があるが、これらに限定されるものではない。また、正孔注入材料に電子受容物質を、電子注入材料に電子供与性物質を添加することにより増感させることもできる。

【００５９】本発明の有機ＥＬ素子において、さらに効果的な電子注入材料は、金属錯体化合物もしくは含窒素五員環誘導体である。具体的には、金属錯体化合物としては、８－ヒドロキシキノリナートリチウム、ビス（８－ヒドロキシキノリナート）亜鉛、ビス（８－ヒドロキシキノリナート）銅、ビス（８－ヒドロキシキノリナート）マンガン、トリス（８－ヒドロキシキノリナート）アルミニウム、トリス（２－メチル－８－ヒドロキシキノリナート）アルミニウム、トリス（８－ヒドロキシキノリナート）ガリウム、ビス（１０－ヒドロキシベンゾ［ｈ］キノリナート）ベリリウム、ビス（１０－ヒドロキシベンゾ［ｈ］キノリナート）亜鉛、ビス（２－メチル－８－キノリナート）クロロガリウム、ビス（２－メチル－８－キノリナート）（ｏ－クレゾラート）ガリウム、ビス（２－メチル－８－キノリナート）（１－ナフトラート）アルミニウム、ビス（２－メチル－８－キノリナート）（２－ナフトラート）ガリウム等があるが、これらに限定されるものではない。また、含窒素五員誘導体としては、オキサゾール、チアゾール、オキサジアゾール、チアジアゾールもしくはトリアゾール誘導体が好ましい。具体的には、２，５－ビス（１－フェニル）－１，３，４－オキサゾール、ジメチルＰＯＰＯＰ、２，５－ビス（１－フェニル）－１，３，４－チアゾール、２，５－ビス（１－フェニル）－１，３，４－オキサジアゾール、２－（４’－ｔｅｒｔ－ブチルフェニル）－５－（４”－ビフェニル）１，３，４－オキサジアゾール、２，５－ビス（１－ナフチル）－１，３，４－オキサジアゾール、１，４－ビス［２－（５－フェニルオキサジアゾリル）］ベンゼン、１，４－ビス［２－（５－フェニルオキサジアゾリル）－４－ｔｅｒｔ－ブチルベンゼン］、２－（４’－ｔｅｒｔ－ブチルフェニル）－５－（４”－ビフェニル）－１，３，４－チアジ

ーチアジアゾール、１，４ービス［２ー（５ーフェニルチアジアゾリル）］ベンゼン、２ー（４’ーｔｅｒｔーブチルフェニル）ー５ー（４”ービフェニル）ー１，３，４ートリアゾール、２，５ービス（１ーナフチル）ー１，３，４ートリアゾール、１，４ービス［２ー（５ーフェニルトリアゾリル）］ベンゼン等があるが、これらに限定されるものではない。

【００６０】本有機ＥＬ素子においては、発光層中に、一般式［１］の化合物の他に、発光材料、ドーピング材料、正孔注入材料および電子注入材料の少なくとも１種が同一層に含有されてもよい。また、本発明により得られた有機ＥＬ素子の、温度、湿度、雰囲気等に対する安定性の向上のために、素子の表面に保護層を設けたり、シリコンオイル、樹脂等により素子全体を保護することも可能である。

【００６１】有機ＥＬ素子の陽極に使用される導電性材料としては、４ｅＶより大きな仕事関数を持つものが適しており、炭素、アルミニウム、バナジウム、鉄、コバルト、ニッケル、タングステン、銀、金、白金、パラジウム等およびそれらの合金、ＩＴＯ基板、ＮＥＳＡ基板に使用される酸化スズ、酸化インジウム等の酸化金属、さらにはポリチオフェンやポリピロール等の有機導電性樹脂が用いられる。陰極に使用される導電性物質としては、４ｅＶより小さな仕事関数を持つものが適しており、マグネシウム、カルシウム、錫、鉛、チタニウム、イットリウム、リチウム、ルテニウム、マンガン、アルミニウム等およびそれらの合金が用いられるが、これらに限定されるものではない。合金としては、マグネシウム／銀、マグネシウム／インジウム、リチウム／アルミニウム等が代表例として挙げられるが、これらに限定されるものではない。合金の比率は、蒸着源の温度、雰囲気、真空度等により制御され、適切な比率に選択される。陽極および陰極は、必要があれば二層以上の層構成により形成されていても良い。

【００６２】有機ＥＬ素子では、効率良く発光させるために、少なくとも一方は素子の発光波長領域において充分透明にすることが望ましい。また、基板も透明であることが望ましい。透明電極は、上記の導電性材料を使用して、蒸着やスパッタリング等の方法で所定の透光性が確保するように設定する。発光面の電極は、光透過率を１０％以上にすることが望ましい。基板は、機械的、熱的強度を有し、透明性を有するものであれば限定されるものではないが、ガラス基板および透明性樹脂フィルムがある。透明性樹脂フィルムとしては、ポリエチレン、エチレンー酢酸ビニル共重合物、エチレンービニルアルコール共重合物、ポリプロピレン、ポリスチレン、ポリメチルメタアクリレート、ポリ塩化ビニル、ポリビニルアルコール、ポリビニルブチラール、ナイロン、ポリエーテルエーテルケトン、ポリサルホン、ポリエーテルサキルビニルエーテル、ポリビニルフルオライド、テトラフルオロエチレンーエチレン、テトラフルオロエチレンーヘキサフルオロプロピレン、ポリクロロトリフルオロエチレン、ポリビニリデンフルオライド、ポリエステル、ポリカーボネート、ポリウレタン、ポリイミド、ポリエーテルイミド、ポリイミド、ポリプロピレン等があげられる。

【００６３】本発明に係わる有機ＥＬ素子の各層の形成は、真空蒸着、スパッタリング、プラズマ、イオンプレーティング等の乾式成膜法やスピンコーティング、ディッピング、フローコーティング等の湿式成膜法のいずれの方法を適用することができる。膜厚は特に限定されるものではないが、適切な膜厚に設定する必要がある。膜厚が厚すぎると、一定の光出力を得るために大きな印加電圧が必要になり効率が悪くなる。膜厚が薄すぎるとピンホール等が発生して、電界を印加しても充分な発光輝度が得られない。通常の膜厚は５ｎｍから１０μｍの範囲が適しているが、１０ｎｍから０．２μｍの範囲がさらに好ましい。

【００６４】湿式成膜法の場合、各層を形成する材料を、エタノール、クロロホルム、テトラヒドロフラン、ジオキサン等の適切な溶媒に溶解または分散させて薄膜を形成するが、その溶媒はいずれであっても良い。また、いずれの有機薄膜層においても、成膜性向上、膜のピンホール防止等のため適切な樹脂や添加剤を使用しても良い。使用の可能な樹脂としては、ポリスチレン、ポリカーボネート、ポリアリレート、ポリエステル、ポリアミド、ポリウレタン、ポリスルフォン、ポリメチルメタクリレート、ポリメチルアクリレート、セルロース等の絶縁性樹脂およびそれらの共重合体、ポリーＮービニルカルバゾール、ポリシラン等の光導電性樹脂、ポリチオフェン、ポリピロール等の導電性樹脂を挙げることができる。また、添加剤としては、酸化防止剤、紫外線吸収剤、可塑剤等を挙げることができる。

【００６５】以上のように、有機ＥＬ素子の発光層に本発明の化合物を用い、更には特定の正孔注入層もしくは電子注入層と組み合わせることにより、発光効率、最大発光輝度等の有機ＥＬ素子特性を改良することができた。また、この素子は熱や電流に対して非常に安定であり、さらには低い駆動電圧で実用的に使用可能の発光輝度が得られるため、従来まで大きな問題であった劣化も大幅に低下させることができた。

【００６６】本発明の有機ＥＬ素子は、壁掛けテレビ等のフラットパネルディスプレイや、平面発光体として、複写機やプリンター等の光源、液晶ディスプレイや計器類等の光源、表示板、標識灯等へ応用が考えられ、その工業的価値は非常に大きい。

【００６７】本発明の材料は、有機ＥＬ素子、電子写真感光体、光電変換素子、太陽電池、イメージセンサー等

【００６８】
【実施例】以下、本発明を実施例に基づきさらに詳細に説明する。
実施例１
洗浄したＩＴＯ電極付きガラス板上に、発光材料として表１の化合物（３）、２，５－ビス（１－ナフチル）－１，３，４－オキサジアゾール、ポリカーボネート樹脂（帝人化成：パンライトＫ－１３００）を５：３：２の重量比でテトラヒドロフランに溶解させ、スピンコーティング法により膜厚１００ｎｍの発光層を得た。その上に、マグネシウムと銀を１０：１で混合した合金で膜厚１５０ｎｍの電極を形成して有機ＥＬ素子を得た。この素子の発光特性は、直流電圧５Ｖで１３０（ｃｄ／m^2）、最高輝度３２００（ｃｄ／m^2）、発光効率０．９５（ｌｍ／Ｗ）の発光が得られた。
【００６９】実施例２
洗浄したＩＴＯ電極付きガラス板上に、表１の化合物（８）を真空蒸着して膜厚１００ｎｍの発光層を作成し、その上に、マグネシウムと銀を１０：１で混合した合金で膜厚１００ｎｍの電極を形成して有機ＥＬ素子を得た。発光層は10^{-6}Ｔｏｒｒの真空中で、基板温度室温の条件下で蒸着した。この素子は、直流電圧５Ｖで１１０（ｃｄ／m^2）、最高輝度２２００（ｃｄ／m^2）、発光効率０．７５（ｌｍ／Ｗ）の発光が得られた。
【００７０】実施例３
洗浄したＩＴＯ電極付きガラス板上に、表１の化合物（１２）を塩化メチレンに溶解させ、スピンコーティング法により膜厚５０ｎｍの発光層を得た。次いで、表３の化合物（Ｂ－１０）を真空蒸着して膜厚３０ｎｍの電子注入層を作成し、その上に、マグネシウムと銀を１０：１で混合した合金で膜厚１００ｎｍの電極を形成して有機ＥＬ素子を得た。発光層および電子注入層は10^{-6}Ｔｏｒｒの真空中で、基板温度室温の条件下で蒸着した。この素子は、直流電圧５Ｖで３５０（ｃｄ／m^2）、最高輝度５４００（ｃｄ／m^2）、発光効率１．３（ｌｍ／Ｗ）の緑色発光が得られた。
【００７１】実施例４
洗浄したＩＴＯ電極付きガラス板上に、表１の化合物（１６）を真空蒸着して、膜厚５０ｎｍに発光層を形成した。次いで、表３の化合物（Ｂ－３）を真空蒸着して膜厚１０ｎｍの電子注入層を作成し、その上に、マグネシウムと銀を１０：１で混合した合金で膜厚１００ｎｍの電極を形成して有機ＥＬ素子を得た。正孔注入層および発光層は10^{-6}Ｔｏｒｒの真空中で、基板温度室温の条件下で蒸着した。この素子は、直流電圧５Ｖで約４１０（ｃｄ／m^2）、最高輝度１００００（ｃｄ／m^2）、発光効率１．６（ｌｍ／Ｗ）の緑色発光が得られた。
【００７２】実施例５～５１
洗浄したＩＴＯ電極付きガラス板上に、表４の条件で、正孔注入材料を真空蒸着して、膜厚３０ｎｍの正孔注入層を得た。次いで、発光材料を真空蒸着して膜厚３０ｎｍの発光層を得た。さらに、電子注入材料を真空蒸着して膜厚３０ｎｍの電子注入層を作成し、その上に、マグネシウムと銀を１０：１で混合した合金で膜厚１５０ｎｍの膜厚の電極を形成して有機ＥＬ素子を得た。各層は10^{-6}Ｔｏｒｒの真空中で、基板温度室温の条件下で蒸着した。この素子の発光特性を表４に示す。ここでの発光輝度は、直流電圧５Ｖ印可時の輝度であり、本実施例の有機ＥＬ素子は、全て最高輝度１００００（ｃｄ／m^2）以上の高輝度特性を有していた。有機ＥＬ素子の素子構成としては、一般式の発光材料に、一般式［３］の正孔注入材料および一般式［４］の電子注入材料を組み合わせた素子が、最も良好な特性を示した。
【００７３】
【表４】

実施例	正孔注入材料	発光材料	電子注入材料	発光輝度 (cd/m²)	最大発光輝度 (cd/m²)	最大発光効率 (lm/W)
5	(A-2)	(8)	(B-3)	4420	33000	4.1
6	(A-8)	(8)	(B-3)	5100	45000	4.2
7	(A-10)	(8)	(B-3)	4800	41000	4.0
8	(A-11)	(8)	(B-3)	5200	38000	3.5
9	(A-12)	(8)	(B-3)	6000	41000	4.0
10	(A-13)	(8)	(B-3)	5800	40000	3.9
11	(A-14)	(8)	(B-3)	6000	41000	3.8
12	(A-16)	(8)	(B-3)	5500	38000	3.5
13	(A-13)	(8)	(B-11)	7700	110000	12.0
14	(A-13)	(1)	(B-11)	4300	55000	7.0
15	(A-13)	(3)	(B-11)	5000	45000	5.9
16	(A-13)	(4)	(B-11)	4400	46000	5.6
17	(A-13)	(7)	(B-11)	4600	51000	5.9
18	(A-13)	(9)	(B-11)	7100	100000	10.0
19	(A-13)	(10)	(B-11)	7600	115000	13.5
20	(A-13)	(11)	(B-11)	7200	104000	10.8
21	(A-13)	(14)	(B-11)	6800	95000	10.1
22	(A-13)	(15)	(B-11)	6500	86000	9.8
23	(A-13)	(16)	(B-11)	7100	115000	13.5
24	(A-13)	(20)	(B-11)	5800	60000	8.8
25	(A-13)	(21)	(B-11)	4900	60000	8.7
26	(A-13)	(23)	(B-11)	6800	75000	7.9
27	(A-13)	(25)	(B-11)	6500	70000	6.9
28	(A-13)	(27)	(B-11)	6800	78000	8.0
29	(A-13)	(34)	(B-11)	6500	81000	8.9
30	(A-13)	(38)	(B-11)	6900	84000	9.1
31	(A-13)	(43)	(B-11)	5800	68000	7.7
32	(A-13)	(45)	(B-11)	6100	62000	7.2
33	(A-13)	(49)	(B-11)	7500	89000	9.9
34	(A-13)	(51)	(B-11)	6000	60000	8.0
35	(A-13)	(53)	(B-11)	7100	95000	9.7
36	(A-13)	(55)	(B-11)	6900	91000	10.5
37	(A-13)	(8)	(B-1)	4500	65000	7.6
38	(A-13)	(8)	(B-4)	4100	58000	6.6
39	(A-13)	(8)	(B-9)	5000	50000	5.9
40	(A-13)	(8)	(B-12)	6900	65000	7.3
41	(A-13)	(8)	(B-13)	7500	106000	11.1
42	(A-13)	(8)	(B-15)	7000	120000	12.6
43	(A-13)	(8)	(B-16)	6200	69000	7.4
44	(A-13)	(8)	(B-18)	5500	70000	6.9
45	(A-13)	(8)	(B-20)	5900	78000	8.0
46	(A-13)	(8)	(B-21)	4300	45000	5.4
47	(A-13)	(8)	(B-22)	4000	32000	4.6
48	(A-13)	(8)	(B-23)	4500	38000	4.3
49	(A-13)	(8)	(B-24)	3300	36000	3.9
50	(A-12)	(8)	(B-14)	6000	81000	8.8
51	(A-8)	(48)	(B-18)	4100	66000	7.2

発光輝度は、素子に直流5（V）印加した時の輝度を示す。

【００７４】実施例５２

洗浄したＩＴＯ電極付きガラス板上に、正孔注入材料（Ａ－１３）を真空蒸着して、膜厚４０ｎｍの正孔注入層を得た。次いで、発光材料として化合物（８）を真空蒸着して膜厚４０ｎｍの発光層を得た。さらに、電子注入材料として（Ｂ－１１）を真空蒸着して、膜厚３０ｎｍの電子注入層を得た。その上に、アルミニウム：リチウムを５０：１の比率の合金で膜厚１５０ｎｍの電極を形成して有機ＥＬ素子を得た。この素子は、直流電圧５Ｖで８０００（ｃｄ／ｍ²）、最高輝度１３１０００（ｃｄ／ｍ²）、発光効率１３．８（ｌｍ／Ｗ）の発光が得られた。

【００７５】実施例５３

ＩＴＯ電極と化合物（Ａ－１３）との間に、無金属フタロシアニンの膜厚５ｎｍの正孔注入層を設ける以外は、の素子は、直流電圧５Ｖで１００００（ｃｄ／ｍ²）、最高輝度９９０００（ｃｄ／ｍ²）、発光効率１０．２（ｌｍ／Ｗ）の発光が得られた。

【００７６】実施例５４

化合物（Ａ－１３）の代わりに無金属フタロシアニンの膜厚１５ｎｍの正孔注入層を設ける以外は、実施例４８と同様の方法で有機ＥＬ素子を作製した。この素子は、直流電圧５Ｖで３５００（ｃｄ／ｍ²）、最高輝度７７０００（ｃｄ／ｍ²）、発光効率５．８（ｌｍ／Ｗ）の発光が得られた。

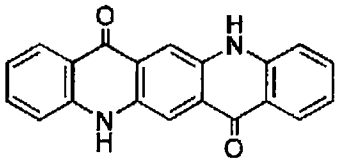
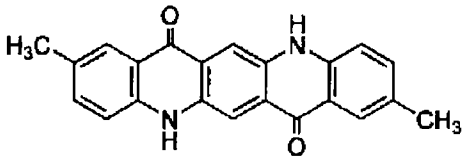
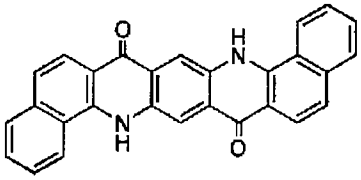
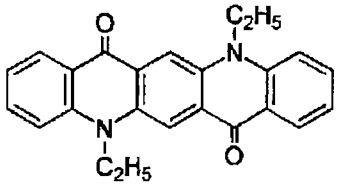
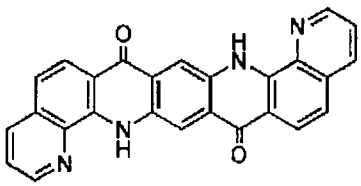
【００７７】実施例５５～６４

発光層として、化合物（８）と表５で示した化合物を１００：１の重量比で蒸着した膜厚２０ｎｍの発光層を使用する以外は、実施例５２と同様の方法で有機ＥＬ素子を作製した。この素子の発光特性を表６に示す。ここで

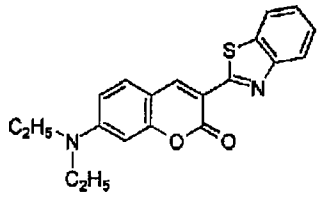
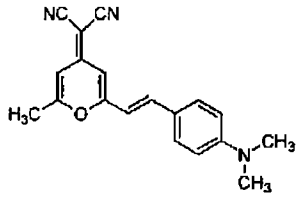
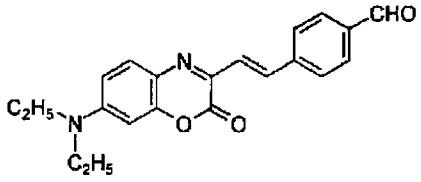
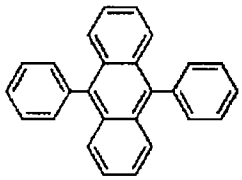
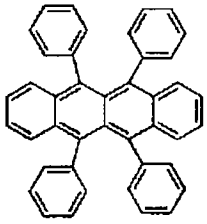
施例の有機ＥＬ素子は、全て最高輝度１００００（ｃｄ／m^2）以上の高輝度特性を有し、また、目的の発光色を得ることができた。

【００７８】

【表５】

化合物	化学構造
Ｃ－１	
Ｃ－２	
Ｃ－３	
Ｃ－４	
Ｃ－５	

【００７９】

化合物	化学構造
C－6	
C－7	
C－8	
C－9	
C－10	

【００８０】

【表６】

実施例	表５のﾄﾞｰﾋﾟﾝｸﾞ材料	発光輝度 (cd/m²)	最大発光輝度 (cd/m²)	最大発光効率 (lm/W)
５５	(C－1)	８３００	１３３０００	１４．０
５６	(C－2)	７６００	１０５０００	１２．８
５７	(C－3)	８５００	１２１０００	１４．１
５８	(C－4)	８２００	１００００００	１０．８
５９	(C－5)	７７００	８９０００	９．９
６０	(C－6)	４３００	５１０００	７．８
６１	(C－7)	５５００	４９０００	８．１
６２	(C－8)	６３００	５５０００	８．９
６３	(C－9)	４８００	５６０００	７．７
６４	(C－10)	５６００	７８０００	７．９

【００８１】本実施例で示された有機ＥＬ素子は、発光輝度として１００００（ｃｄ／ｍ²）以上であり、全て高い発光効率を得ることができた。本実施例で示された有機ＥＬ素子について、３（ｍＡ／ｃｍ²）で連続発光させたところ、１０００時間以上安定な発光を観測した。本発明の有機ＥＬ素子材料を使用した有機ＥＬ素子は、発光材料の蛍光量子効率が極めて高いので、この発光材料を使用した素子においては、低電流印可領域での高輝度発光が可能になり、また、発光層中で一般式［１］の化合物に加えてドーピング材料を使用すること

らには、青緑色、緑色および黄色の発光をする一般式［1］の化合物に、赤色発光もしくは青色発光のドーピング材料を添加することによって、赤色発光もしくは青色発光の発光素子を得ることができた。

【００８２】本発明の有機ＥＬ素子は発光効率、発光輝度の向上と長寿命化を達成するものであり、併せて使用される発光材料、ドーピング材料、正孔注入材料、電子注入材料、増感剤、樹脂、電極材料等および素子作製方法を限定するものではない。

【００８３】

【発明の効果】本発明の有機ＥＬ素子材料を発光材料として使用した有機ＥＬ素子は、従来に比べて高い発光効率で高輝度の発光を示し、長寿命の有機ＥＬ素子を得ることができた。以上により本発明で示した化合物を、有機ＥＬ素子の少なくとも一層に使用すること、および、本発明の素子構成により形成された有機ＥＬ素子は、高輝度、高発光効率、長寿命の有機ＥＬ素子を容易に作製することが可能となった。

PATENT ABSTRACTS OF JAPAN

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(21)Application number : 08-078501 (71)Applicant : TOYO INK MFG CO LTD

(22)Date of filing : 01.04.1996 (72)Inventor : ENOKIDA TOSHIO

TAMANO MICHIKO

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(54) LUMINESCENT MATERIAL FOR ORGANIC ELECTROLUMINESCENCE ELEMENT AND ORGANIC ELECTROLUMINESCENT ELEMENT USING THE SAME

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a luminescent material for organic electroluminescent element which has a specific chemical structure, can attain high intensity and luminescent properties of high efficiency, shows excellent durability in stability, when used repetitively, and is useful in a high-intensity luminescent element to be used in a planar light source or a display.

SOLUTION: This luminescent material has a chemical structure represented by formula I {A and X1-X4 are each a (substituted) 6-20C arylene; Y1-Y4 are each a group of formula II [R1-R4 are each H, (substituted) alkyl, (substituted) aryl, cyano; R1 and R2 or R3 and R4 may bond to each other to form a triple bond; Z is a (substituted) aryl; n is 0, 1]; a-d are each 0-2} and typically N,N,N', N'-tetrakis(4-styrilphenyl)-p-phenylenediamine is exemplified.

LEGAL STATUS [Date of request for examination] 02.04.2001

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[Kind of final disposal of application other than the examiner's decision of rejection or application converted registration]

[Date of final disposal for application]

[Patent number] 3564859

[Date of registration] 18.06.2004

[Number of appeal against examiner's decision of rejection]

[Date of requesting appeal against examiner's decision of rejection]

[Date of extinction of right]

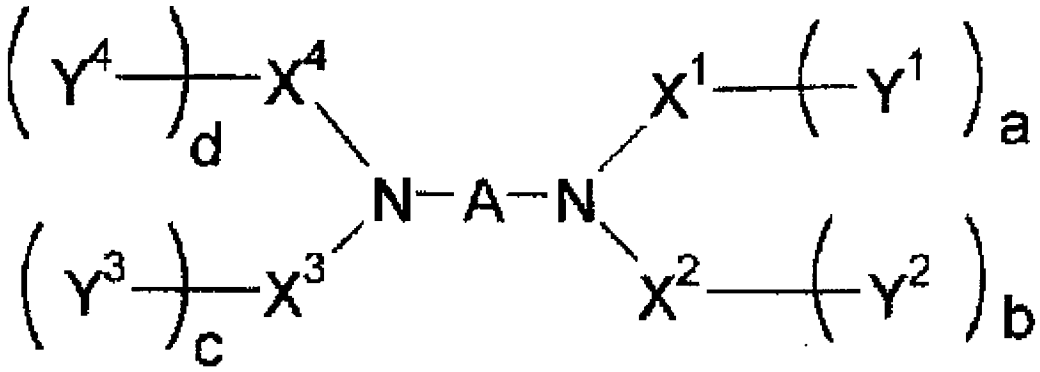
CLAIMS

[Claim(s)]

[Claim 1] The charge of organic electroluminescent element material shown by the following general formula [1].

General formula [1]

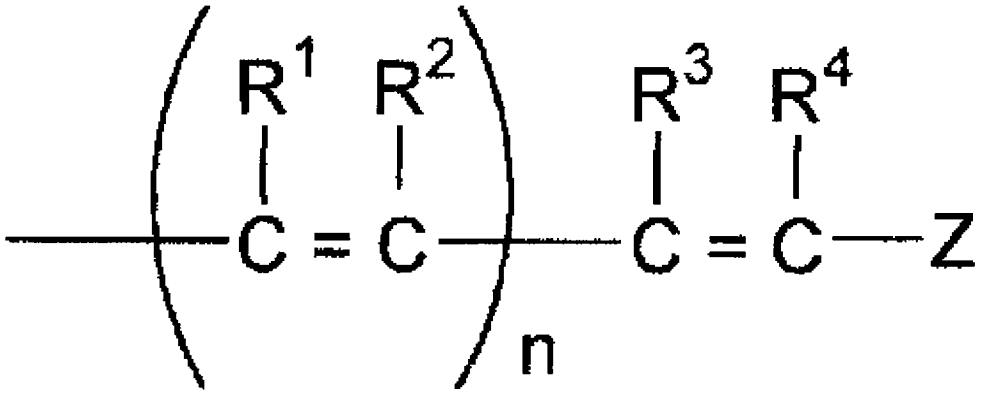
[Formula 1]



A and X1 -X4 express independently the arylene radical of the carbon atomic numbers 6-20 which are not permuted [a permutation or] among [type, respectively. Y1 -Y4 The organic radical shown by the following general formula [2] is expressed independently, respectively.

General formula [2]

[Formula 2]



(R1 -R4 expresses among a formula the triple bond which expressed the aryl group which is not permuted [the alkyl group which is not permuted / a hydrogen atom, a permutation, or /, a permutation, or] and the cyano group, or R1, R2, or R3 and R4 combined.) Z expresses the aryl group which is not permuted [a permutation or]. a-d expresses the positive integer of 0-2. n expresses 0 or 1.]

[Claim 2] The charge of organic electroluminescent element material according to claim 1 whose A is the condensation arylene radical of the carbon atomic numbers 10-20 which are not permuted [a permutation or] in the above-mentioned general formula [1].

[Claim 3] The charge of organic electroluminescent element material according to claim 1 or 2 which is the luminescent material for organic electroluminescent elements.

[Claim 4] The organic electroluminescent element which is the layer in which at least one layer contains claim 1 thru/or the charge of organic electroluminescent

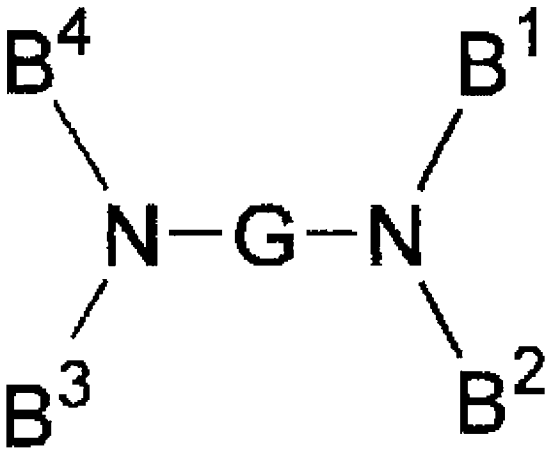
element material given in three in the organic electroluminescent element which comes to form the organic compound thin film of two or more layers which contains a luminous layer or a luminous layer in inter-electrode [of a pair].

[Claim 5] The organic electroluminescent element according to claim 4 which comes to form the layer containing the third class amine derivative of aromatic series, and/or a phthalocyanine derivative between a luminous layer and an anode plate.

[Claim 6] The organic electroluminescent element according to claim 5 the third class amine derivative of whose of aromatic series is the compound shown by the following general formula [3].

General formula [3]

[Formula 3]



B1 - B4 express independently the aryl group of the carbon atomic numbers 6-20 which are not permuted [a permutation or] among [type, respectively. G

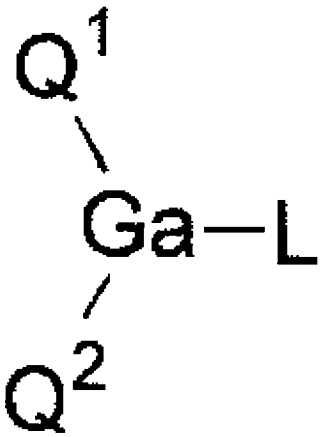
expresses the arylene radical which is not permuted [a permutation or].]

[Claim 7] Claim 4 which comes to form the layer containing a metal complex compound or a nitrogen-containing five membered ring derivative between a luminous layer and cathode thru/or an organic electroluminescent element given in six.

[Claim 8] Claim 4 whose metal complex compound is a compound shown by the following general formula [4] thru/or the organic electroluminescent element of seven publications.

General formula [4]

[Formula 4]



Q1 and Q2 express independently the hydroxy benzoquinoline derivative which is not permuted [the hydroxyquinoline derivative which is not permuted / a permutation or /, a permutation, or] among [type, respectively. L The cycloalkyl radical which is not permuted [the alkyl group which is not permuted / a halogen

atom, a permutation, or /, a permutation, or], The aryl group, -OR (R is the aryl group which may also contain the nitrogen atom which is not permuted [the cycloalkyl radical which is not permuted / the alkyl group which is not permuted / a hydrogen atom, a permutation, or /, a permutation, or /, a permutation, or].) which may also contain the nitrogen atom which is not permuted [a permutation or] The ligand expressed with -O-Ga-Q3 (Q(Q4) 3 and Q4 express the same semantics as Q1 and Q2.) is expressed.]

DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the luminescent material for the organic (electroluminescence EL) components and the light emitting device of high brightness which are used for the flat-surface light source or a display.

[0002]

[Description of the Prior Art] Promising ** of the application as a large area full color display device with a cheap solid-state luminescence mold is carried out, and, as for the EL element which used the organic substance, many

development is performed. Generally EL consists of counterelectrodes of the pair the luminous layer and this whose layer were pinched. When electric field are impressed between two electrodes, an electron is poured in from a cathode side and, as for luminescence, an electron hole is poured in from an anode plate side. Furthermore, in case this electron recombines with an electron hole in a luminous layer and an energy level returns from a conduction band to a valence band, it is the phenomenon which emits energy as a light.

[0003] Compared with the inorganic EL element, the conventional organic EL device had high driver voltage, and luminescence brightness and its luminous efficiency were also low. Moreover, property degradation is also remarkable and it had not resulted in utilization. In recent years, the organic EL device which carried out the laminating of the thin film containing an organic compound with the high fluorescence quantum efficiency which emits light by the low battery not more than 10V is reported, and the interest is attracted (refer to applied FIJIKUSU Letters, 51 volumes, 913 pages, and 1987). This approach used the luminous layer and the amine system compound for the hole injection layer for the metal chelate complex, green luminescence of high brightness has been obtained, several 1000 cd/m2 and the maximum luminous efficiency attain 1.5 lm/W with the direct current voltage of 6-7V, and brightness has the engine performance near a practical use field.

[0004] However, although luminescence reinforcement is improved for the organic EL device to current by the improvement of a configuration, it does not have still sufficient luminescence brightness. Moreover, it has the big problem of being inferior to the stability at the time of repeat use. This had chemically unstable metal chelate complexes, such as for example, a tris(8-hydroxyquinolinate)aluminium complex, at the time of electroluminescence, that of adhesion with cathode was bad, and had deteriorated greatly in short-time luminescence. Development of the luminescent material which has the luminescence capacity which was excellent for development of the organic EL device which had high luminescence brightness and luminous efficiency and was excellent in the stability in the time of repeat use, and is durable for the above reason is desired.

[0005]

[Problem(s) to be Solved by the Invention] Luminescence brightness of this invention is high and it is in offer of the organic EL device which was excellent in the stability in the time of repeat use. as a result of this invention persons' inquiring wholeheartedly, the luminescence brightness and luminous efficiency of an organic EL device which boiled further the charge of organic EL device material shown by the general formula [1], and used it at least are high, and it finds out that the stability in the time of repeat use is also excellent, and came to

accomplish this invention.

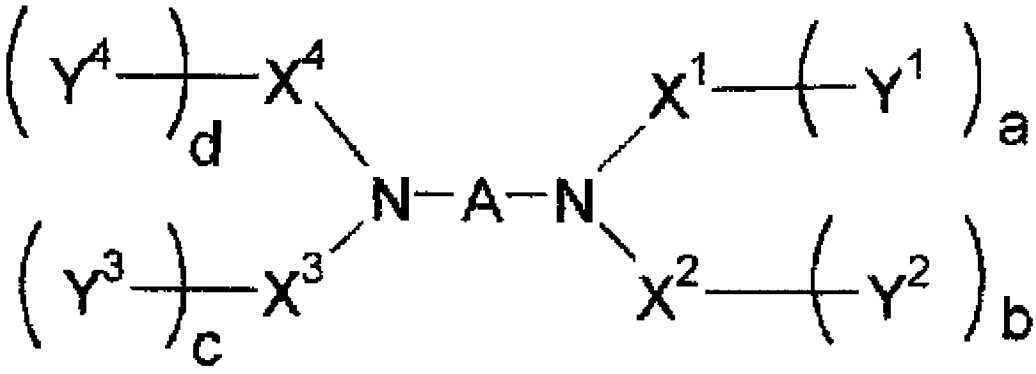
[0006]

[Means for Solving the Problem] This invention relates to the charge of organic electroluminescent element material shown by the following general formula [1].

General formula [1]

[0007]

[Formula 5]

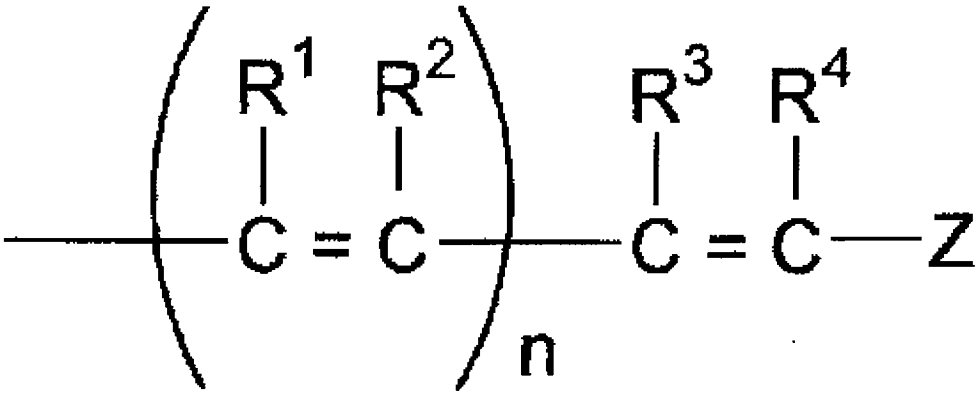


[0008] A and X1 -X4 express independently the arylene radical of the carbon atomic numbers 6-20 which are not permuted [a permutation or] among [type, respectively. Y1 -Y4 The organic radical shown by the following general formula [2] is expressed independently, respectively.

General formula [2]

[0009]

[Formula 6]



[0010] (R1 -R4 expresses among a formula the triple bond which expressed the aryl group which is not permuted [the alkyl group which is not permuted / a hydrogen atom, a permutation, or /, a permutation, or] and the cyano group, or R1, R2, or R3 and R4 combined.) Z expresses the aryl group which is not permuted [a permutation or]. a-d expresses the positive integer of 0-2. n expresses 0 or 1.]

[0011] Furthermore, this invention is the above-mentioned charge of organic electroluminescent element material whose A is the condensation arylene radical of the carbon atomic numbers 10-20 which are not permuted [a permutation or] in the above-mentioned general formula [1].

[0012] Furthermore, this invention is a luminescent material for organic electroluminescent elements shown by the above-mentioned general formula [1].

[0013] Furthermore, this invention is an organic electroluminescent element which is the layer in which at least one layer contains the above-mentioned

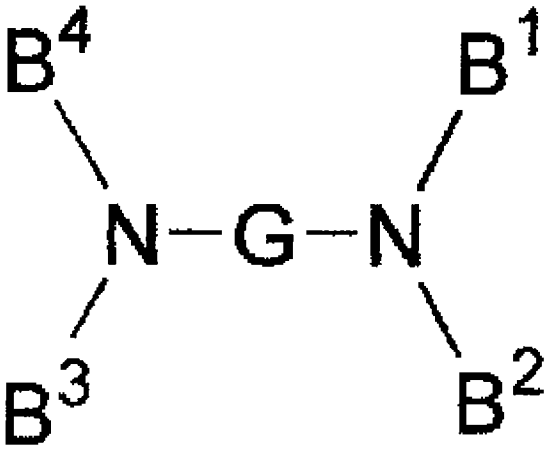
charge of organic electroluminescent element material in the organic electroluminescent element which comes to form the organic compound thin film of two or more layers which contains a luminous layer or a luminous layer in inter-electrode [of a pair].

[0014] Furthermore, this invention is the above-mentioned organic electroluminescent element which comes to form the layer containing the third class amine derivative of aromatic series, and/or a phthalocyanine derivative between a luminous layer and an anode plate.

[0015] Furthermore, this invention is the above-mentioned organic electroluminescent element the third class amine derivative of whose of aromatic series is the compound shown by the following general formula [3].

General formula [3]

[Formula 7]



B1 - B4 express independently the aryl group of the carbon atomic numbers

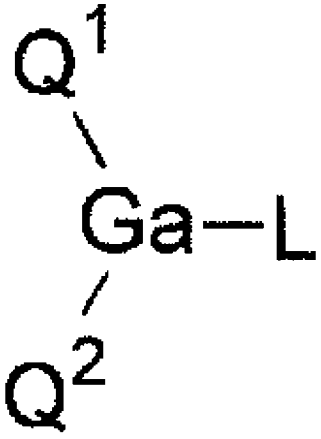
6-20 which are not permuted [a permutation or] among [type, respectively. G expresses the arylene radical which is not permuted [a permutation or].]

[0016] Furthermore, this invention is the above-mentioned organic electroluminescent element which comes to form the layer containing a metal complex compound or a nitrogen-containing five membered ring derivative between a luminous layer and cathode.

[0017] Furthermore, this invention is the above-mentioned organic electroluminescent element whose metal complex compound is a compound shown by the following general formula [4].

General formula [4]

[Formula 8]



Q1 and Q2 express independently the hydroxy benzoquinoline derivative which is not permuted [the hydroxyquinoline derivative which is not permuted / a permutation or /, a permutation, or] among [type, respectively. L The cycloalkyl

radical which is not permuted [the alkyl group which is not permuted / a halogen atom, a permutation, or /, a permutation, or], The aryl group, -OR (R is the aryl group which may also contain the nitrogen atom which is not permuted [the cycloalkyl radical which is not permuted / the alkyl group which is not permuted / a hydrogen atom, a permutation, or /, a permutation, or /, a permutation, or].) which may also contain the nitrogen atom which is not permuted [a permutation or] The ligand expressed with -O-Ga-Q3 (Q(Q4) 3 and Q4 express the same semantics as Q1 and Q2.) is expressed.]

[Embodiment of the Invention]

[0018] A of the compound shown by the general formula [1] in this invention, and X1 -X4 The arylene radical of the carbon atomic numbers 6-20 which are not permuted [a permutation or] is expressed independently, respectively. A and X1 -X4 Although an example is an arylene radical shown by an arylene radical and the following general formula [5], or general formulas [6], such as a phenylene group, a biphenylene radical, a terphenylene radical, a naphthylene radical, anthrylene group, a phenan tolylene radical, a full ORENIREN radical, a pyrenylene radical, and a thio phenylene group, it is not limited to these. Moreover, the condensation arylene radical of the carbon atomic numbers 10-20 expresses the fused aromatic ring of the bivalence which consists of carbon atomic numbers 10-20 like a naphthylene radical, anthrylene group, a phenan

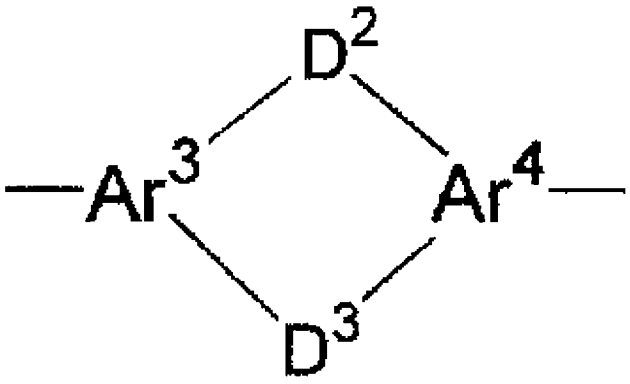
tolylene radical, a full ORENIREN radical, and a pyrenylene radical.

[0019] General formula [5]

[Formula 9]

[0020] General formula [6]

[Formula 10]



[0021] here -- Ar1 -Ar4 of a general formula [5] and a general formula [6] The arylene radical of the carbon atomic numbers 6-20 which are not permuted [a permutation or] is expressed independently, respectively. The examples of Ar1 -Ar 4 are arylene radicals, such as a phenylene group, a biphenylene radical, a terphenylene radical, a naphthylene radical, anthrylene group, a phenan tolylene radical, a full ORENIREN radical, a pyrenylene radical, and a thio phenylene group. D1 - D3 They are direct coupling or the joint radical of bivalence which

consists of 1-50 atoms chosen from C, N, H, O, and S and which consists of rational combination chemically.

[0022] R1 -R4 of the compound shown by the general formula [2] in this invention The aryl group or cyano group which is not permuted [the alkyl group which is not permuted / a hydrogen atom, a permutation, or /, a permutation, or] is expressed independently, respectively. R1 -R4 An example as an alkyl group which is not permuted [a permutation or] A methyl group, an ethyl group, a propyl group, butyl, sec-butyl, tert-butyl, A pentyl radical, a hexyl group, a heptyl radical, an octyl radical, a stearyl radical, 2-phenyl isopropyl group, a trichloromethyl radical, a trifluoromethyl radical, There are benzyl, alpha-phenoxybenzyl radical, alpha, and alpha-dimethylbenzyl radical, alpha, and alpha-methylphenyl benzyl, alpha, and alpha-ditrifluoromethyl benzyl, a triphenylmethyl radical, an alpha-benzyloxybenzyl radical, etc. As an aryl group which is not permuted [a permutation or], there are a phenyl group, 2-methylphenyl radical, 3-methylphenyl radical, 4-methylphenyl radical, 4-ethyl phenyl group, a biphenyl radical, 4-methyl biphenyl radical, 4-ethyl biphenyl radical, 4-cyclohexyl biphenyl radical terphenyl radical, 3, 5-dichlorophenyl radical, a naphthyl group, 5-methyl naphthyl group, an anthryl radical, a pyrenyl radical, etc. In a general formula [1], a-d expresses the positive integer of 0-2 independently, respectively, and n expresses 0 or 1 independently, respectively.

[0023] Z of the compound shown by the general formula [2] in this invention expresses independently the aryl group of the carbon atomic numbers 6-20 which are not permuted [a permutation or], respectively. The examples of Z are aryl groups, such as a phenyl group, a biphenyl radical, a terphenyl radical, a naphthyl group, an anthryl radical, a phenan tolyl group, a fluorenyl group, a pyrenyl radical, and a thiophene radical, and the above-mentioned aryl group may have the substituent. The example of a substituent is R1 -R4. In addition to the alkyl group and aryl group which were described, there are an alkoxy group, the amino group, a cyano group, a hydroxyl group, a carboxylic-acid radical, a ether group, an ester group, etc.

[0024] Although the example of representation of the compound of the general formula [1] of this invention is concretely illustrated to Table 1 below, this invention is not limited to this example of representation.

[0025]

[Table 1]

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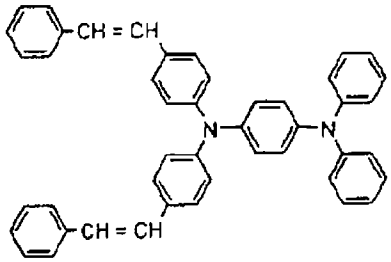
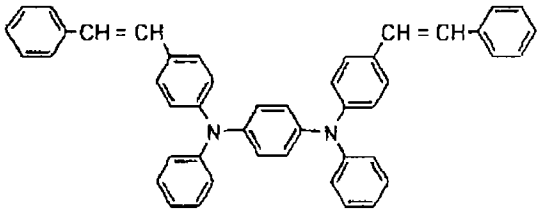
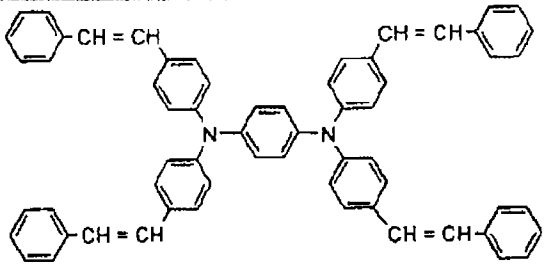
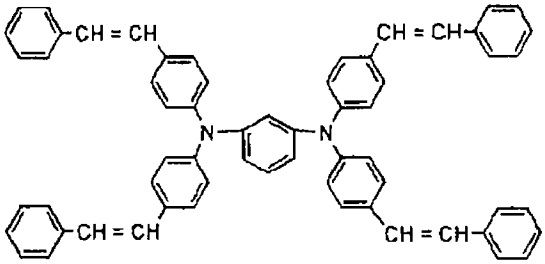
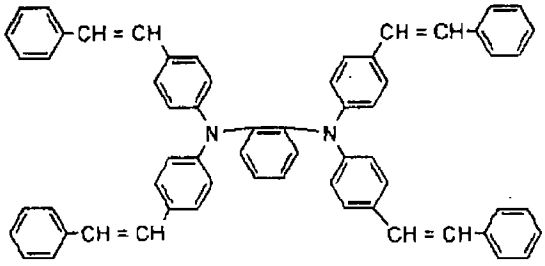
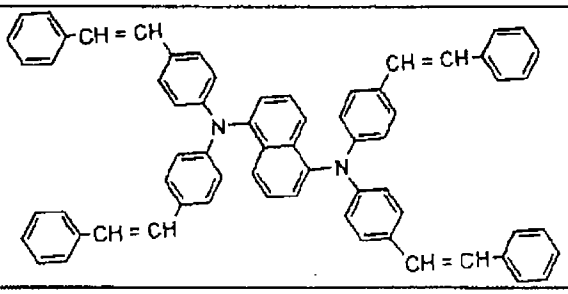
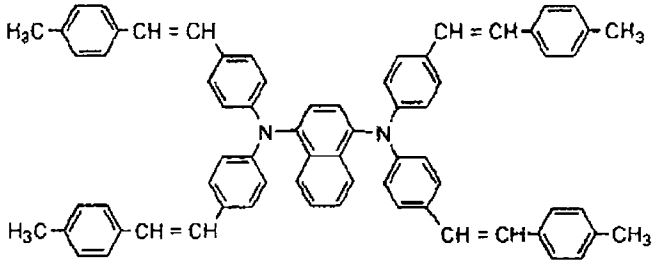
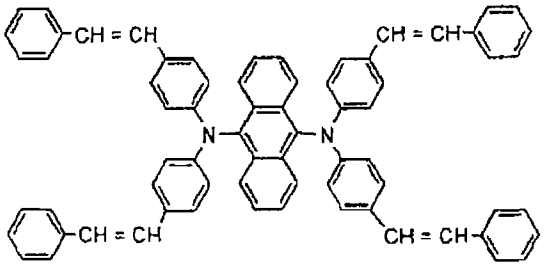
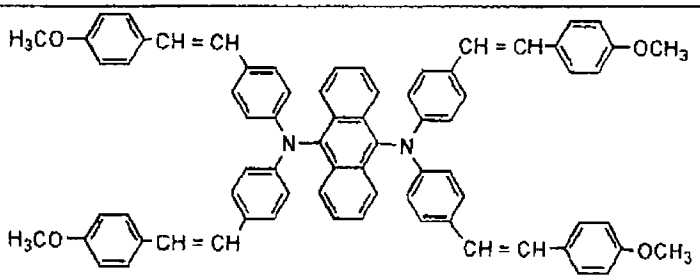
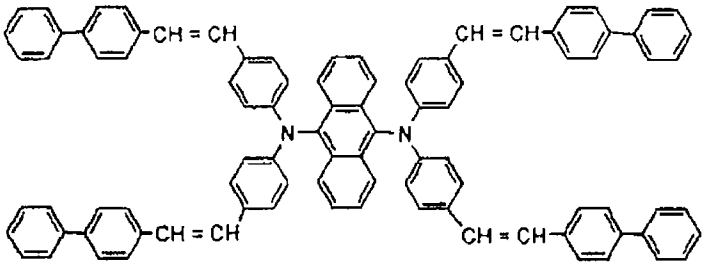
(1)	
(2)	
(3)	
(4)	
(5)	

[0026]

化合物	化 学 構 造 式
(6)	
(7)	
(8)	
(9)	
(10)	

[0027]

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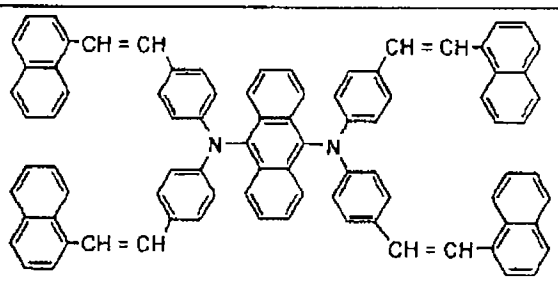
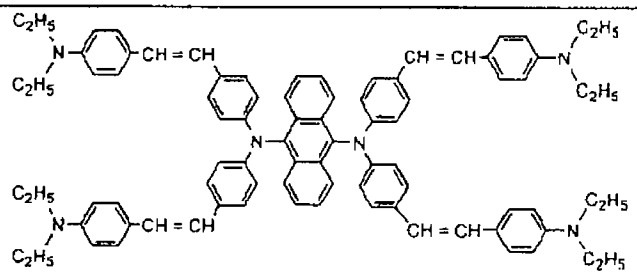
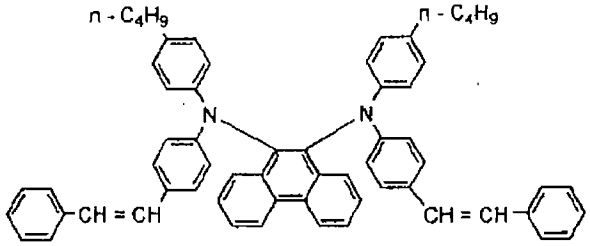
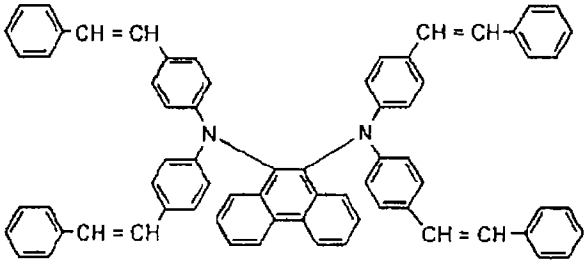
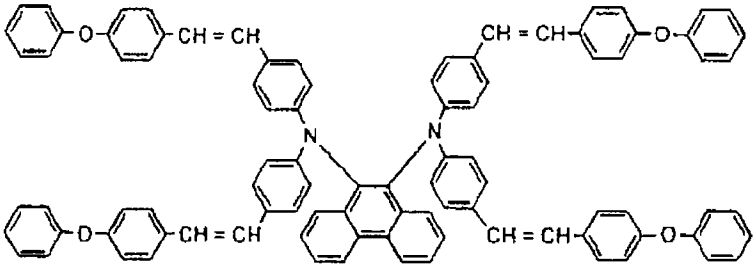
（１１）	
（１２）	
（１３）	
（１４）	
（１５）	

[0028]

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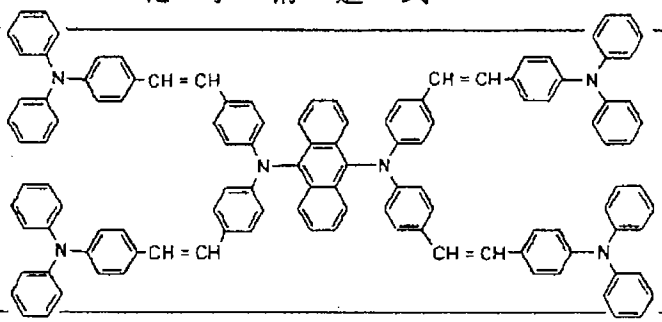
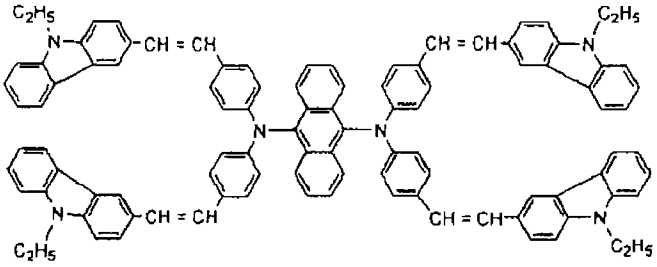
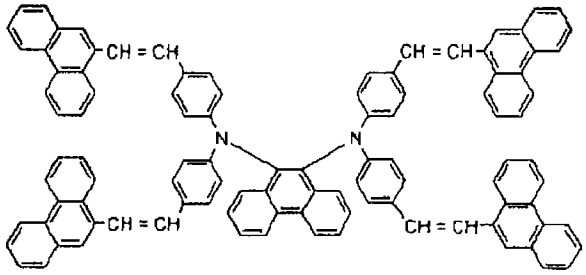
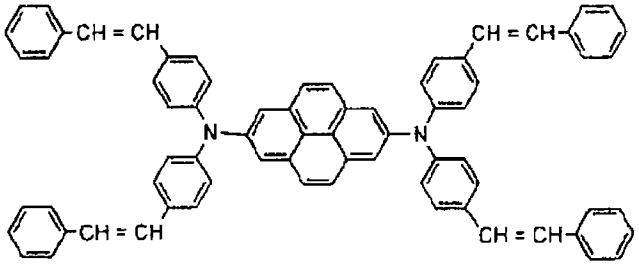
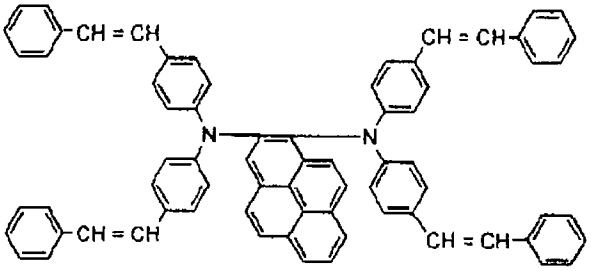
（１６）	
（１７）	
（１８）	
（１９）	
（２０）	

[0029]

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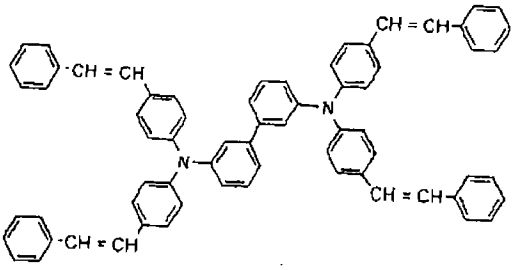
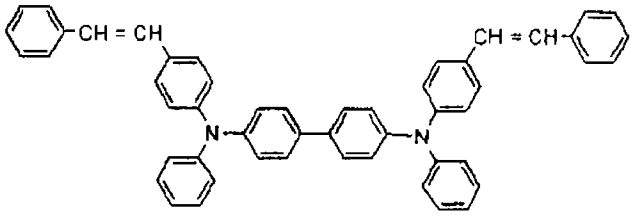
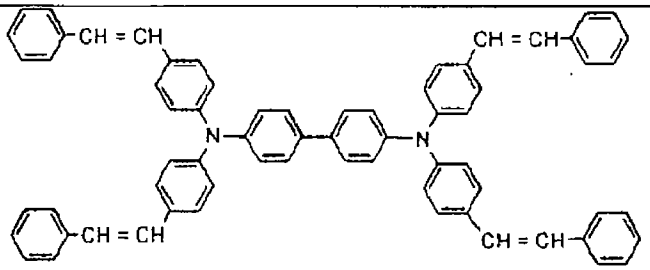
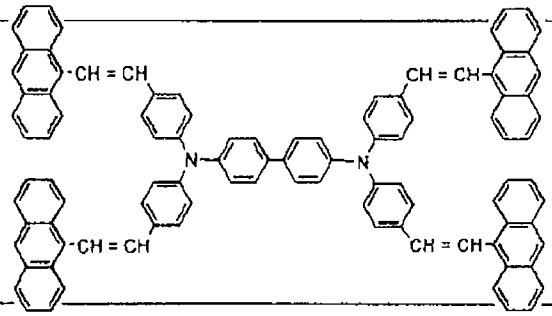
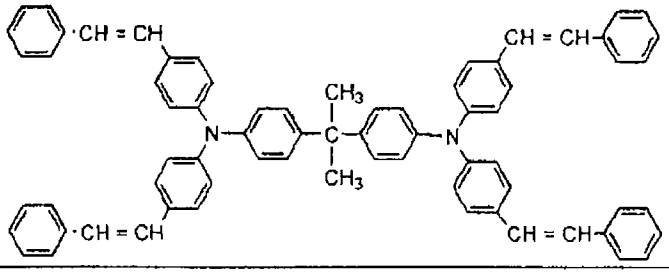
（２１）	
（２２）	
（２３）	
（２４）	
（２５）	

[0030]

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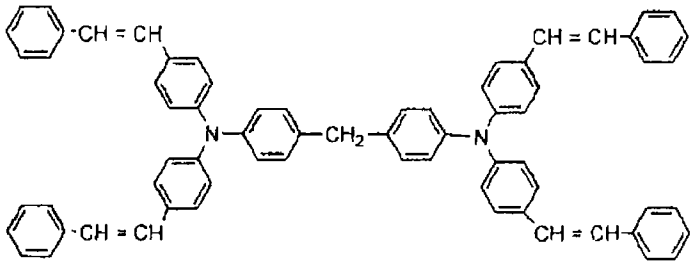
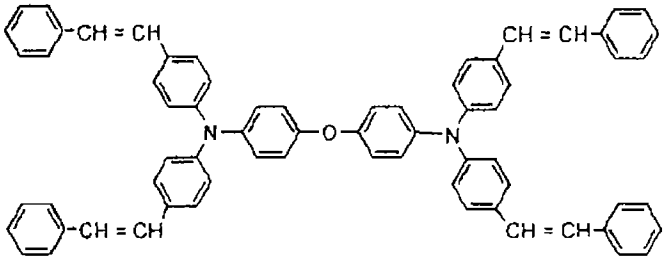
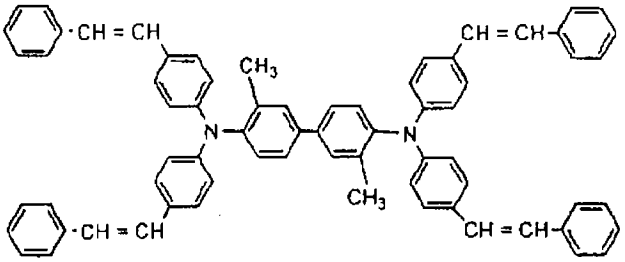
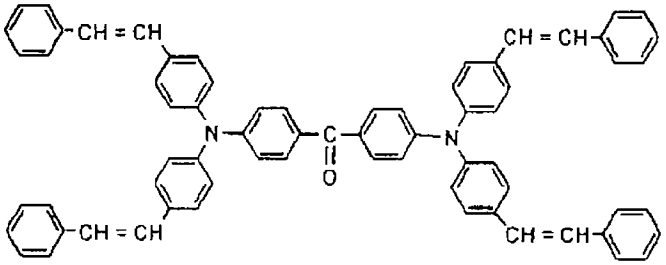
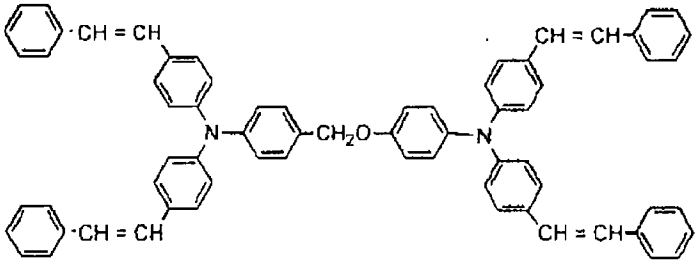
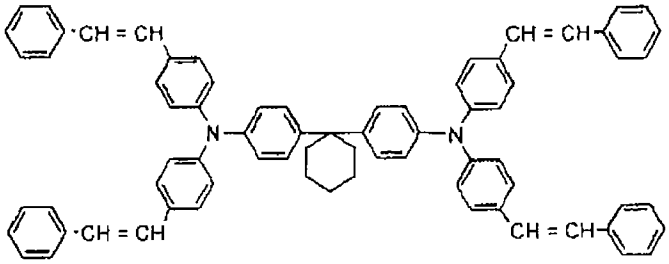
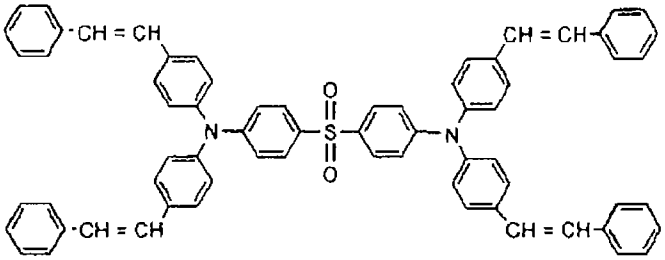
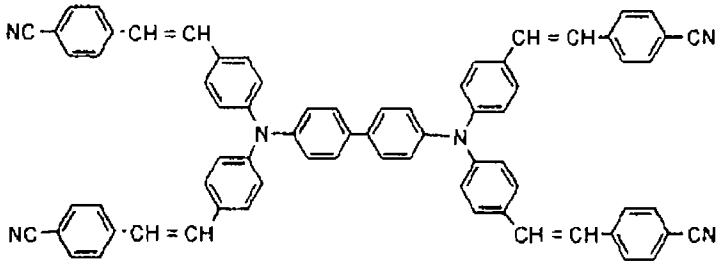
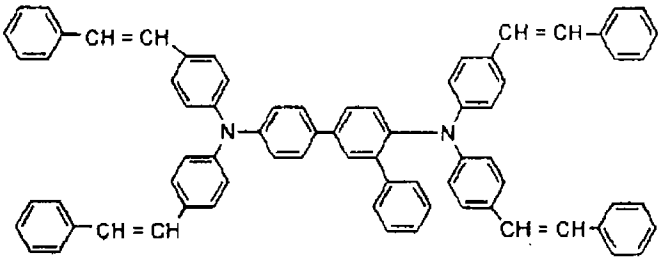
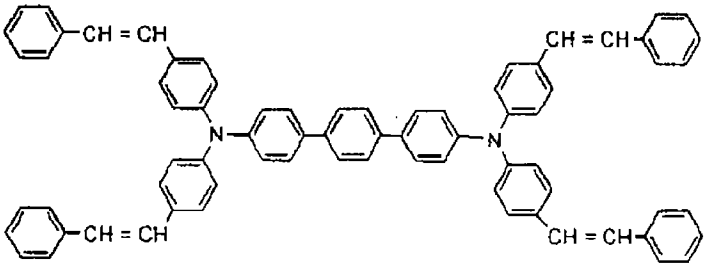
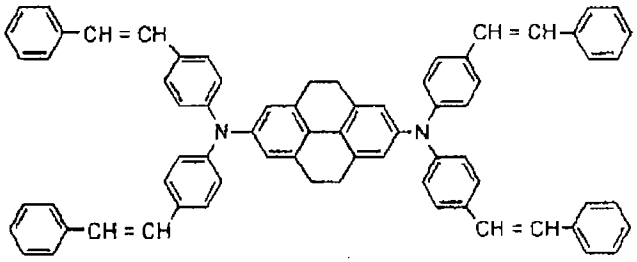
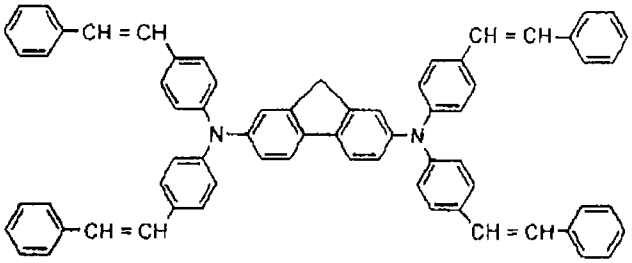
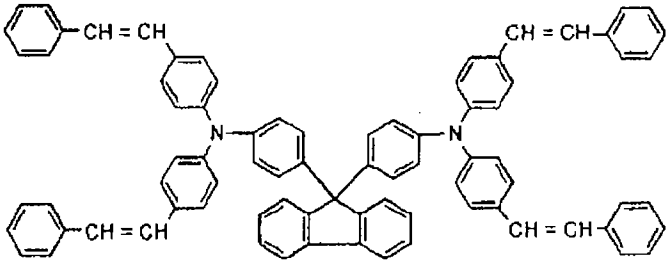
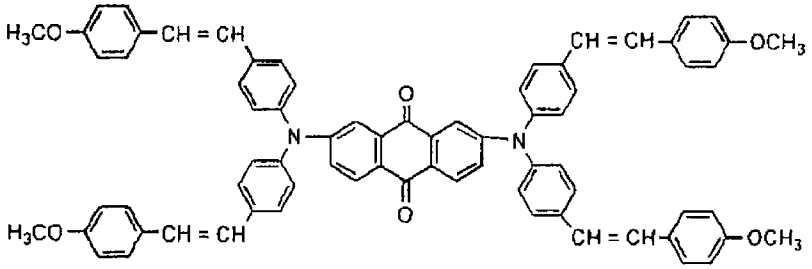
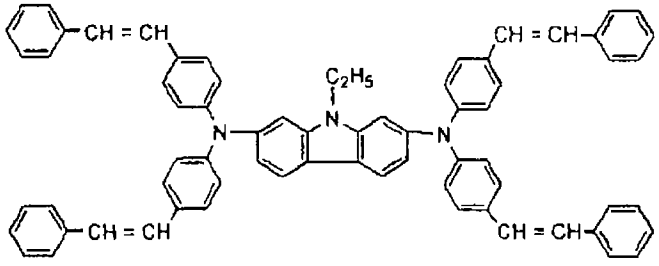
（２６）	
（２７）	
（２８）	
（２９）	
（３０）	

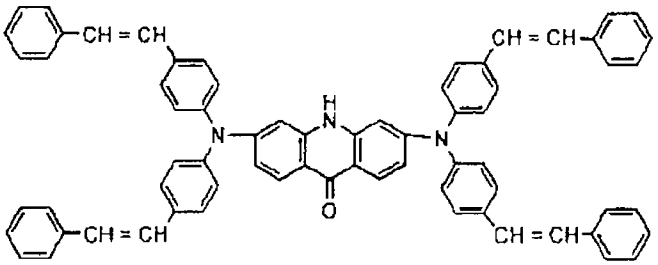
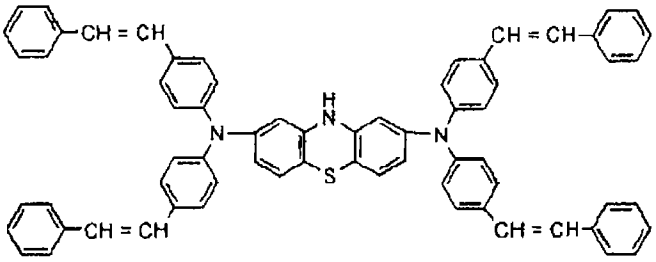
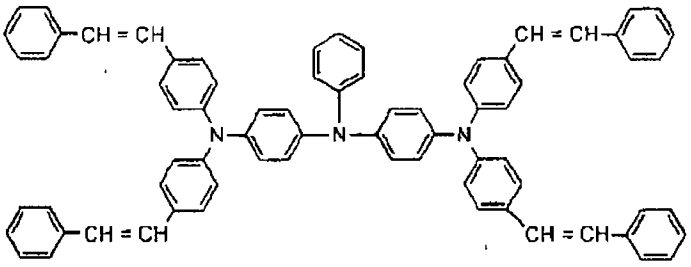
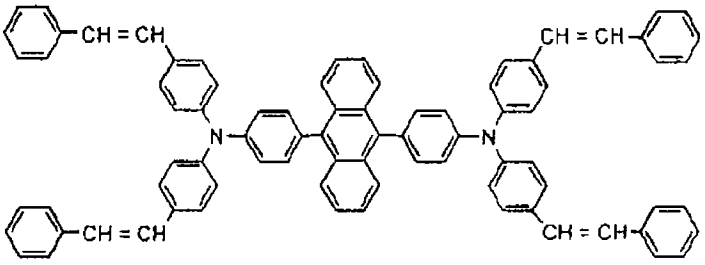
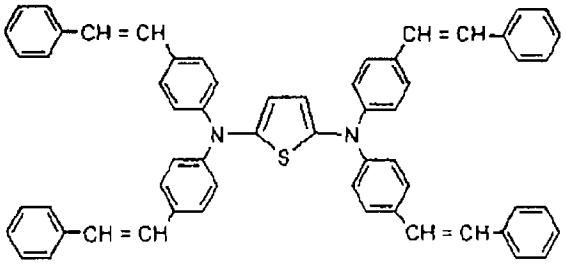
[0031]

化合物	化学構造式
(31)	
(32)	
(33)	
(34)	
(35)	

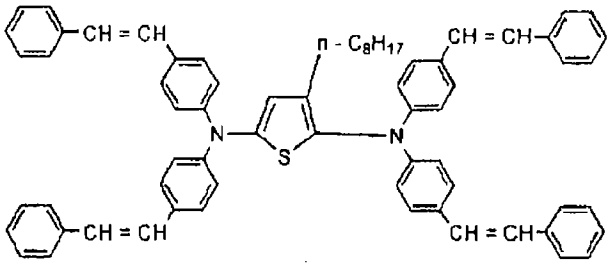
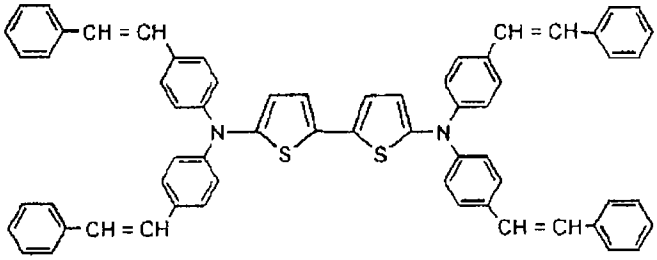
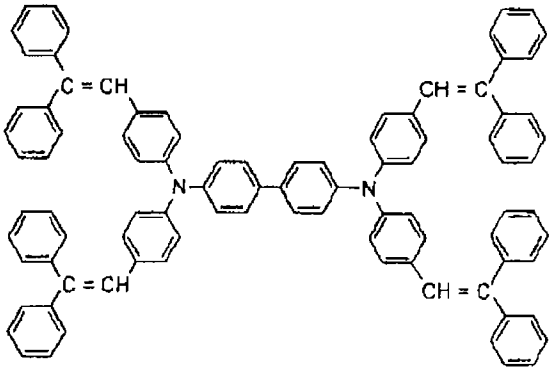
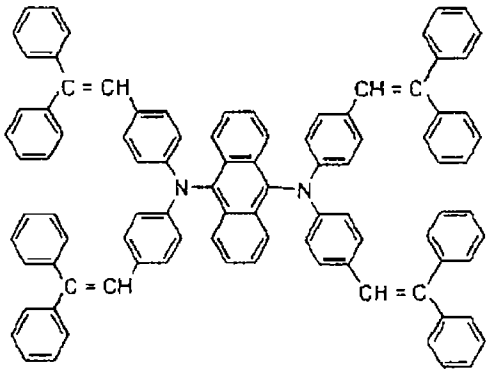
[0032]

化合物	化 学 構 造 式
(36)	
(37)	
(38)	
(39)	
(40)	

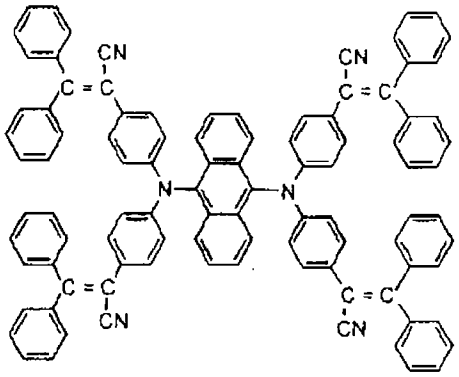
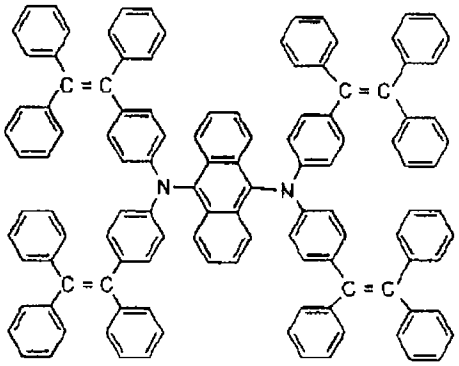
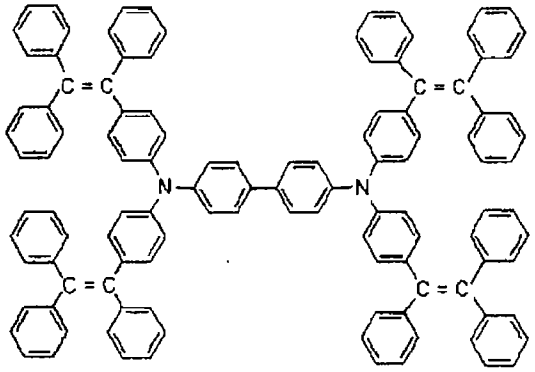
[0033]

化合物	化 学 構 造 式
(41)	
(42)	
(43)	
(44)	
(45)	

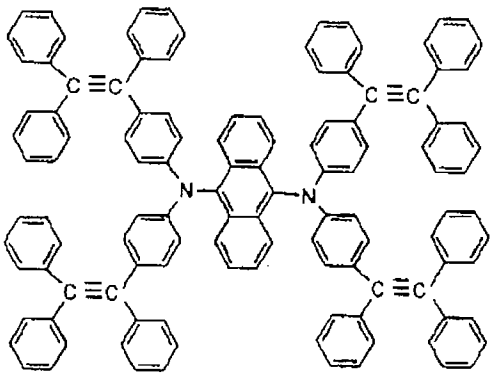
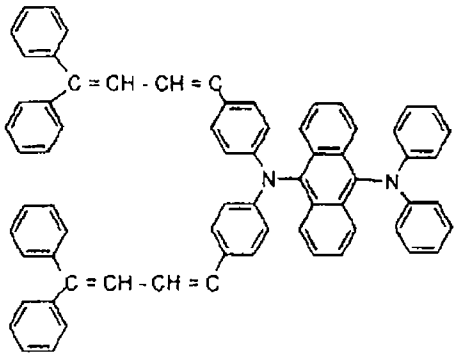
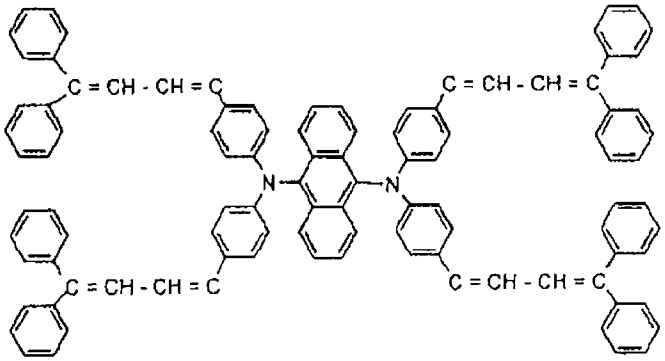
[0034]

化合物	化 学 構 造 式
(46)	
(47)	
(48)	
(49)	

[0035]

化合物	化 学 構 造 式
(50)	
(51)	
(52)	

[0036]

化合物	化 学 構 造 式
(53)	
(54)	
(55)	

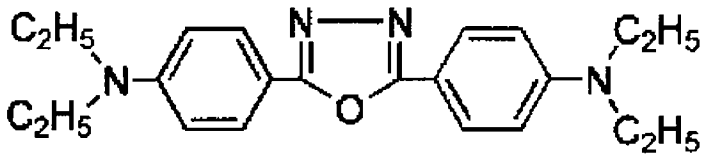
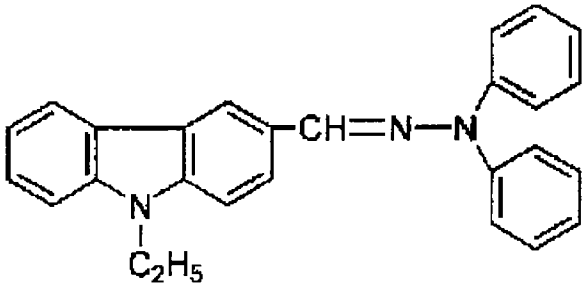
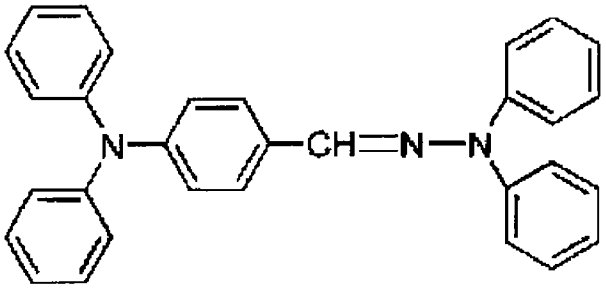
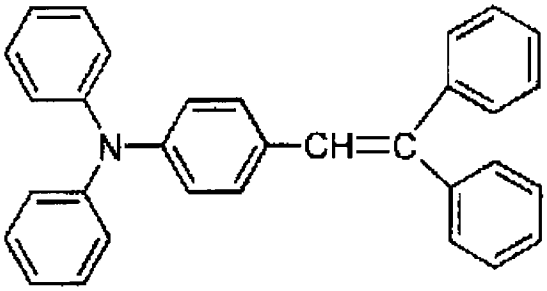
[0037] B1 of the compound shown by the general formula [3] in this invention - B4 An example is the aryl group of the carbon atomic numbers 6-20 which are not permuted [a permutation or]. It is the aryl group which may contain nitrogen atoms, such as a phenyl group, a biphenyl radical, a terphenyl radical, a

naphthyl group, an anthryl radical, a phenan tolyl group, a fluorenyl group, and a pyrenyl radical, and, specifically, each aryl group may have the substituent. G is the arylene radical of bivalence, it is the arylene radical of the bivalence which may contain nitrogen atoms, such as a phenylene group, a biphenylene radical, a terphenylene radical, a naphthylene radical, anthrylene group, a phenan tolylene radical, a full ORENIREN radical, and a pyrenylene radical, and each aryl group may have the substituent.

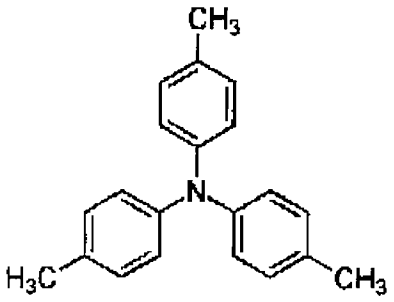
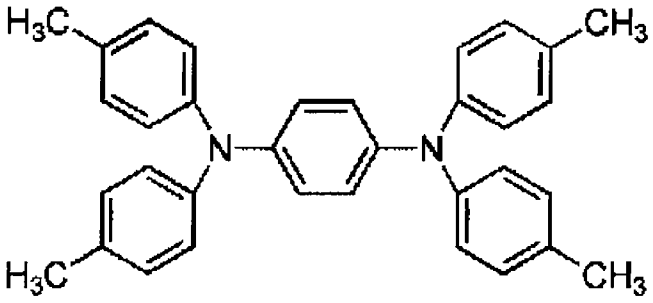
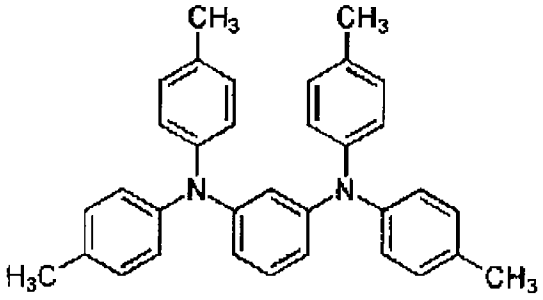
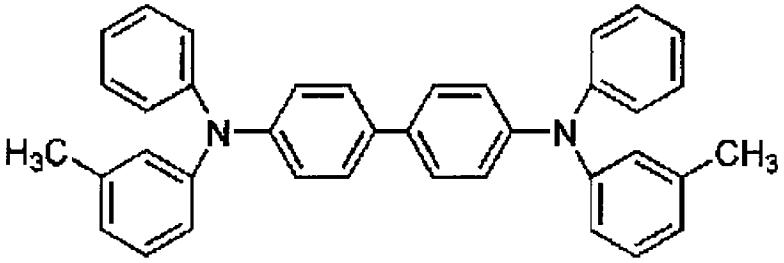
[0038] Although the example of representation of the compound of the general formula [3] of this invention which is a hole-injection ingredient effective for below, and other ingredients is concretely illustrated to Table 2, this invention is not limited to this example of representation.

[0039]

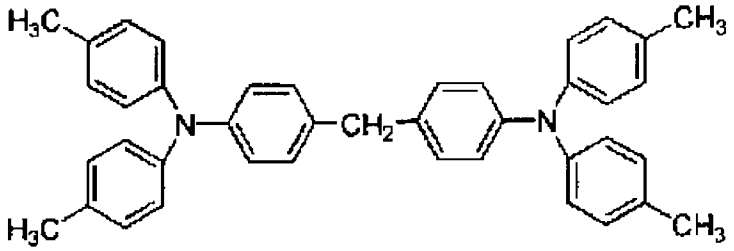
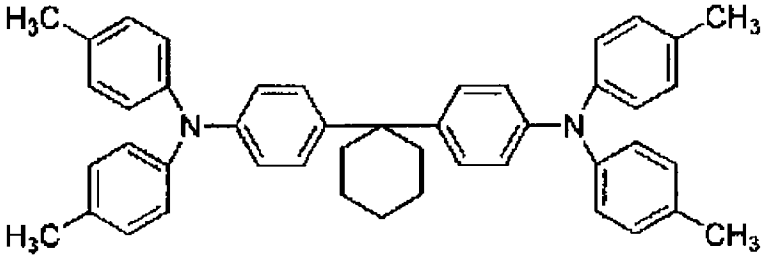
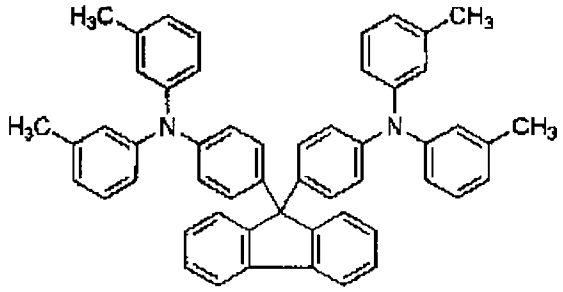
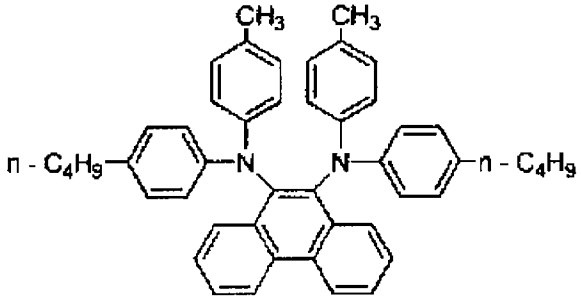
[Table 2]

化合物	化　学　構　造
A－1	
A－2	
A－3	
A－4	

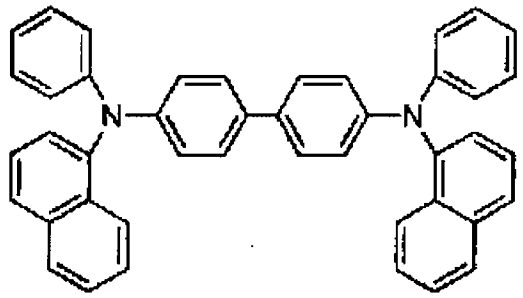
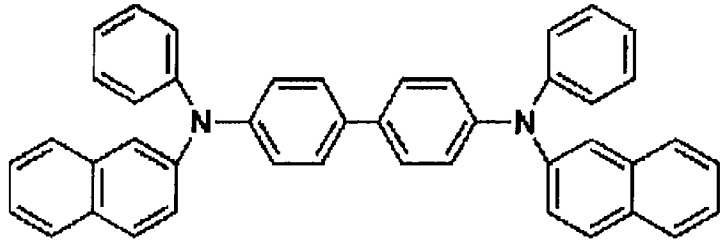
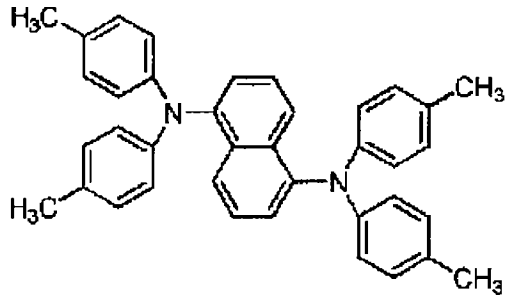
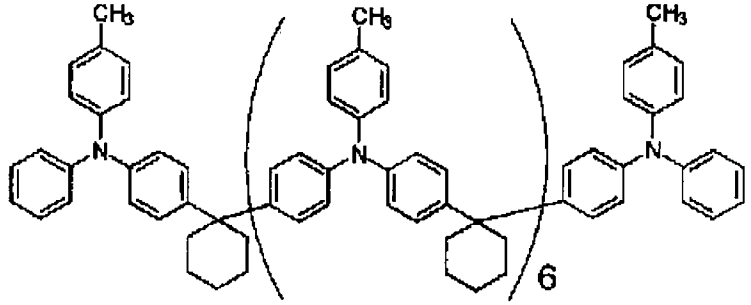
[0040]

化合物	化　学　構　造
A－5	
A－6	
A－7	
A－8	

[0041]

化合物	化　学　構　造
A－9	
A－10	
A－11	
A－12	

[0042]

化合物	化 学 構 造
A－13	
A－14	
A－15	
A－16	

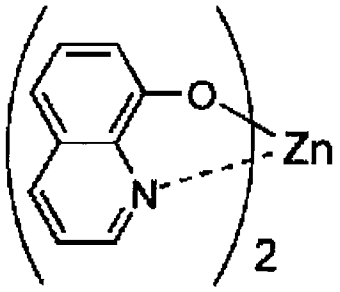
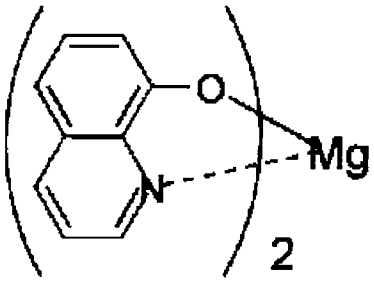
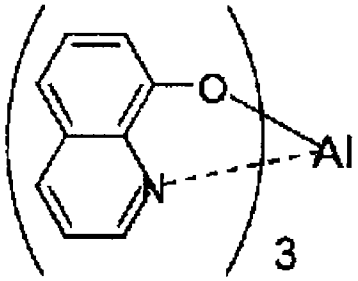
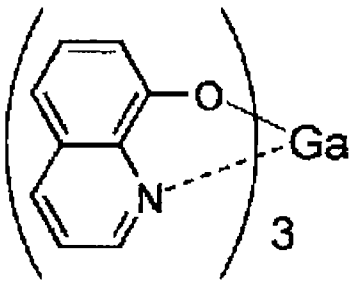
[0043] Q1 of the compound shown by the general formula [4] in this invention, and Q4 8-hydroxyquinoline, a 8-hydroxy quinaldine, a 8-hydroxy-2-phenyl

quinoline, A 8-hydroxy-5-methyl quinoline, 8-hydroxy - hydroxyquinoline derivatives, such as 3, 5, and 7-trifluoro quinoline, and L The cycloalkyl radical which is not permuted [the alkyl group which is not permuted / a halogen atom, a permutation, or /, a permutation, or], The aryl group, -OR (R is the aryl group which may also contain the nitrogen atom which is not permuted [the cycloalkyl radical which is not permuted / the alkyl group which is not permuted / a hydrogen atom, a permutation, or /, a permutation, or /, a permutation, or].) which may also contain the nitrogen atom which is not permuted [a permutation or] -O-Ga-Q3 (Q(Q4) 3 and Q4 express the same semantics as Q1 and Q2.) is shown. Here, the alkyl group of R of a halogen atom, an alkyl group, a cycloalkyl radical, the aryl group that may also contain a nitrogen atom, and -OR radical, a cycloalkyl radical, and the aryl group that may also contain a nitrogen atom are R1 -R4 described by the aforementioned general formula [2]. The same radical is expressed.

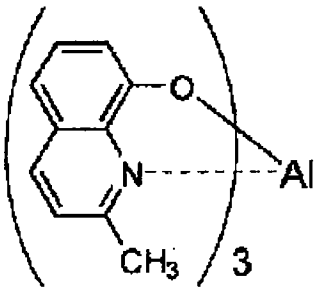
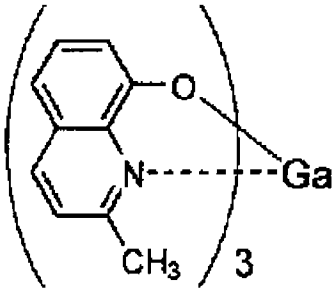
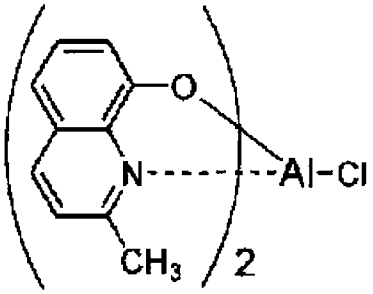
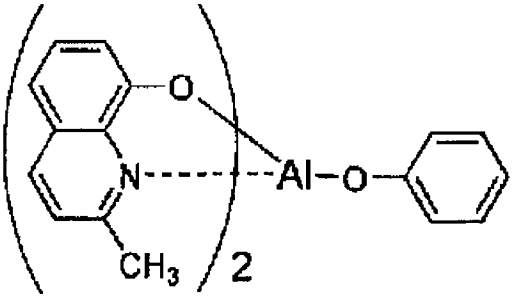
[0044] Although the example of representation of the compound of the general formula [4] used for the organic EL device of this invention below and the example of representation of an electron injection ingredient are concretely illustrated to Table 3, this invention is not limited to this example of representation.

[0045]

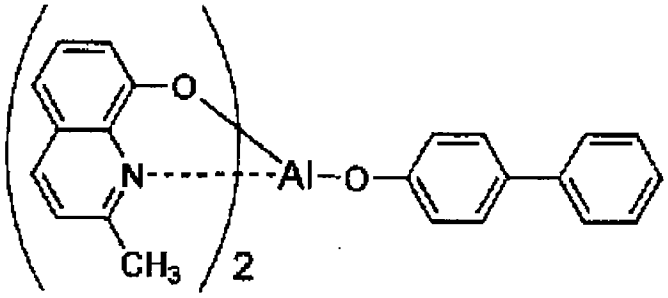
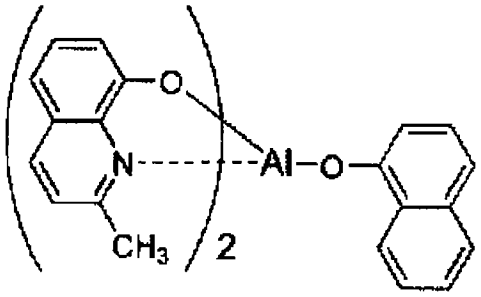
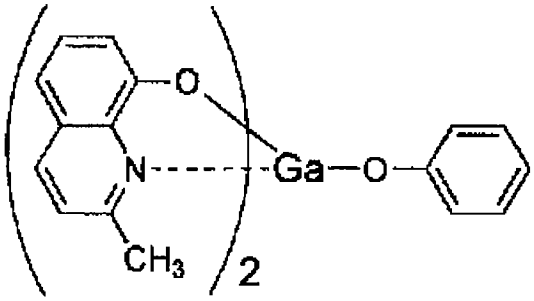
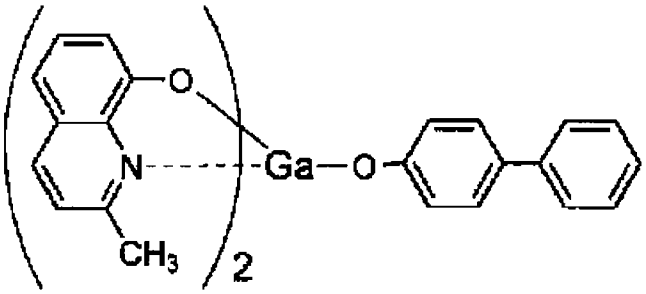
[Table 3]

化合物	化　学　構　造
B－1	
B－2	
B－3	
B－4	

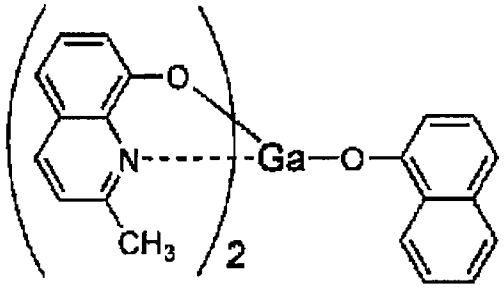
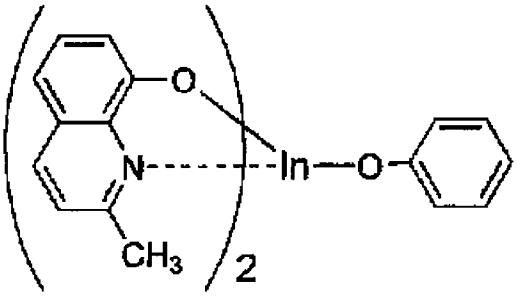
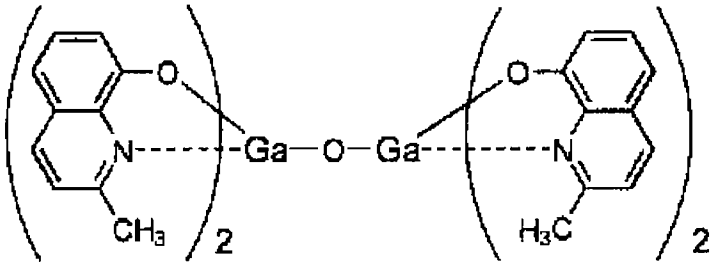
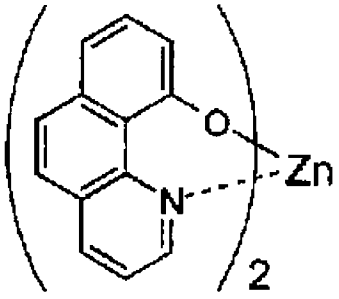
[0046]

化合物	化　学　構　造
B－5	
B－6	
B－7	
B－8	

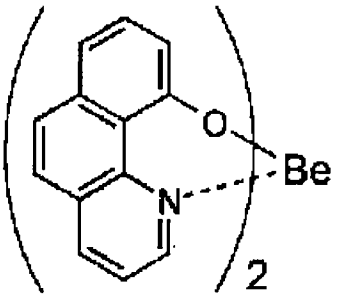
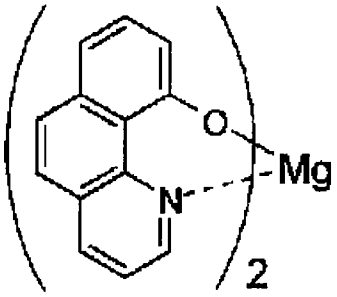
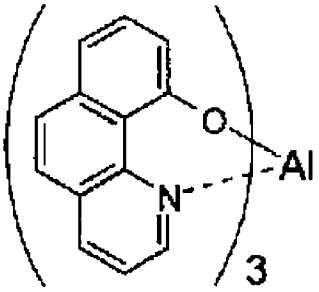
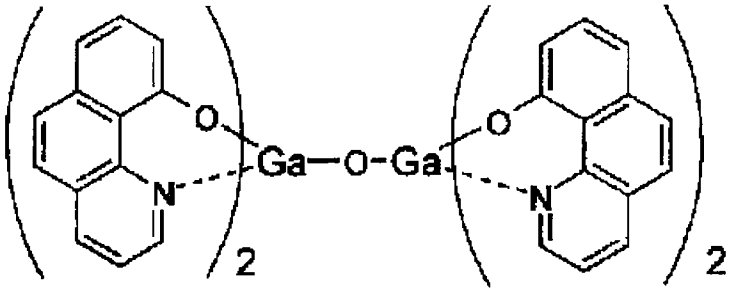
[0047]

化合物	化　学　構　造
B－9	
B－10	
B－11	
B－12	

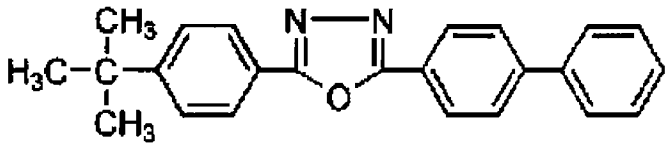
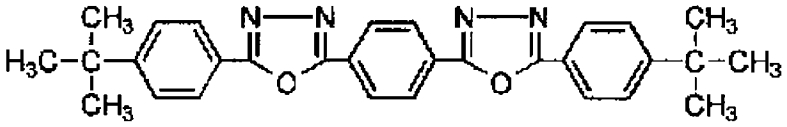
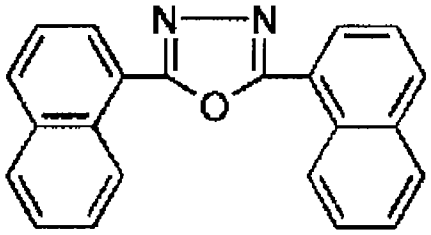
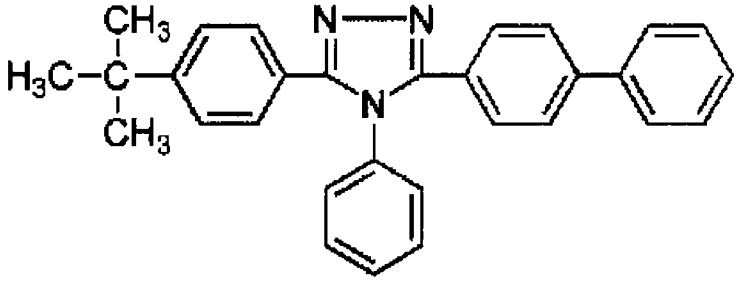
[0048]

化合物	化　学　構　造
B－１３	
B－１４	
B－１５	
B－１６	

[0049]

化合物	化　学　構　造
B－１７	
B－１８	
B－１９	
B－２０	

[0050]

化合物	化　学　構　造
B－２１	
B－２２	
B－２３	
B－２４	

[0051] The compound shown by the general formula [1] of this invention is a compound which has strong fluorescence in a solid state, and is excellent also in

electroluminescence nature. Moreover, since it has collectively the electron injection nature which was excellent from hole injectional [which was excellent from the metal electrode or the organic thin film layer] and electron hole transportability, the metal electrode, or the organic thin film layer, and electronic transportability with it, it can be effectively used as a luminescent material, and even if it uses the electron hole transportability ingredient, electronic transportability ingredient, or doping ingredient of further others, it does not interfere.

[0052] An organic EL device is a component in which one layer or a multilayer organic thin film was formed between an anode plate and cathode. In the case of the mold, the luminous layer is further prepared between an anode plate and cathode. A luminous layer contains luminescent material, and in order to make the electron hole which was poured in from the anode plate in addition to it, or the electron poured in from cathode convey to luminescent material, it may contain a hole-injection ingredient or an electron injection ingredient. However, since the luminescent material of this invention has very high luminescence quantum efficiency, high electron hole transport capacity, and electronic transport capacity and can form a uniform thin film, it can also form a luminous layer only by the luminescent material of this invention. A multilayer mold has the organic EL device which carried out the laminating with the multilayer

configuration of (an anode plate / hole injection layer / luminous layer / cathode), (an anode plate / luminous layer / electronic injection layer / cathode), and (an anode plate / hole injection layer / luminous layer / electronic injection layer / cathode). Since the compound of a general formula [1] has a high luminescence property and has hole injectional, electron hole transport properties and electron injection nature, and electronic transport properties, it can be used for a luminous layer as a luminescent material.

[0053] If there is need, in addition to the compound of the general formula [1] of this invention, the further well-known luminescent material, a doping ingredient, a hole-injection ingredient, and an electron injection ingredient can also be used for a luminous layer. An organic EL device can prevent the fall of the brightness by quenching, or a life by making it multilayer structure. If there is need, it can be used combining luminescent material, a doping ingredient, a hole-injection ingredient, or an electron injection ingredient. Moreover, improvement in luminescence brightness or luminous efficiency, and red and blue luminescence can also be obtained with a doping ingredient. Moreover, a hole injection layer, a luminous layer, and an electronic injection layer may be formed of the lamination more than a bilayer, respectively. In the case of a hole injection layer, in that case, the layer which conveys [the layer which pours in an electron hole from an electrode] an electron hole for an electron hole from a hole injection layer and a

hole injection layer to a reception luminous layer is called an electron hole transportation layer. Similarly, in the case of an electronic injection layer, the layer which conveys [the layer which pours in an electron from an electrode] an electron for an electron from an electronic injection layer and an electronic injection layer to a reception luminous layer is called an electron transport layer. These each class is used by each factor, such as adhesion with the energy level of an ingredient, thermal resistance, an organic layer, or a metal electrode, choosing it.

[0054] As the luminescent material which can be used for a luminous layer with the compound of a general formula [1], or a doping ingredient An anthracene, naphthalene, a phenanthrene, a pyrene, tetracene, Coronene, a chrysene, a fluorescein, perylene, phtalo perylene, Non [naphthalo perylene and peri non, / phtalo peri non, / naphthalo peri] A diphenyl butadiene, a tetra-phenyl butadiene, a coumarin, oxadiazole, Aldazine, bis-benzoKISAZORIN, bis-styryl, pyrazine, a cyclopentadiene, A quinoline metal complex, an amino quinoline metal complex, a benzoquinoline metal complex, Although there are an imine, diphenylethylene, a vinyl anthracene, a diamino carbazole, a pyran, thiopyran, poly methine, merocyanine, an imidazole chelation oxy-NOIDO compound, Quinacridone, rubrene, a fluorochrome, etc., it is not limited to these.

[0055] The compound which has the capacity to convey an electron hole, as a

hole-injection ingredient, has the hole-injection effectiveness which was excellent to the hole-injection effectiveness, the luminous layer, or luminescent material from an anode plate, and prevented migration into the electronic injection layer or electron injection ingredient of an exciton generated by the luminous layer, and was excellent in the thin film organization potency force is mentioned. Specifically A phthalocyanine derivative, a naphthalocyanine derivative, a porphyrin derivative, Oxazole, oxadiazole, triazole, an imidazole, imidazolone, Imidazole thione, pyrazoline, a pyrazolone, a tetrahydro imidazole, Oxazole, oxadiazole, a hydrazone, an acyl hydrazone, The poly aryl alkane, a stilbene, a butadiene, a benzidine mold triphenylamine, a styryl amine mold triphenylamine, a diamine mold triphenylamine, etc., Although there are polymeric materials, such as those derivatives and a polyvinyl carbazole, polysilane, and a conductive polymer, etc., it is not limited to these.

[0056] The still more effective hole-injection ingredient in the hole-injection ingredient which can be used in the organic EL device of this invention is the third class amine derivative of aromatic series or phthalocyanine derivative shown by the general formula [3]. Specifically A triphenylamine, a tritolyl amine, a tolyl diphenylamine, N, N'-diphenyl-N, N' - (3-methylphenyl) -1, the 1'-biphenyl -4, 4'-diamine, N, N, N', N' - (4-methylphenyl) -1, the 1'-phenyl -4, 4'-diamine, N, N, N', N' - (4-methylphenyl) -1, the 1'-biphenyl -4, 4'-diamine, N and N' -

diphenyl-N and N' - dinaphthyl -1 and 1' -- the - biphenyl -4 and 4' - diamine -- N, N'-(methylphenyl)-N, N' -(4-n-buthylphenyl)- A phenanthrene -9, 10-diamine, Although there is oligomer or a polymer with the third class amines frame of such aromatic series, such as an N and N-bis(4-G 4-tolylamino phenyl)-4-phenyl-cyclohexane, etc., it is not limited to these.

[0057] As a phthalocyanine (Pc) derivative, although there are a phthalocyanine derivative, naphthalocyanine derivatives, etc., such as H2 Pc, CuPc, CoPc, NiPc, ZnPc, PdPc, FePc, MnPc, ClAlPc, ClGaPc, ClInPc, ClSnPc, Cl2 SiPc, (HO) AlPc, (HO) GaPc, VOPc, TiOPc, MoOPc, and GaPc-O-GaPc, it is not limited to these.

[0058] The compound which has the capacity to convey an electron, as an electron injection ingredient, has the electron injection effectiveness which was excellent to the hole-injection effectiveness, the luminous layer, or luminescent material from cathode, and prevented migration to the hole injection layer of the exciton generated by the luminous layer, and was excellent in the thin film organization potency force is mentioned. for example, full -- me -- non, although there are anthra quinodimethan, diphenoquinone, thiopyran dioxide, oxazole, oxadiazole, triazole, an imidazole, perylene tetracarboxylic acid, deflection ORENIRIDEN methane, anthra quinodimethan, anthrones, etc. and those derivatives, it is not limited to these. Moreover, sensitization can be carried out by adding the electronic acceptance matter into a hole-injection ingredient, and

adding the electron-donative matter into an electron injection ingredient.

[0059] In the organic EL device of this invention, a still more effective electron injection ingredient is a metal complex compound or a nitrogen-containing five membered ring derivative. As a metal complex compound, specifically 8-hydroxyquinolinate lithium, Bis(8-hydroxyquinolinate)zinc, bis(8-hydroxyquinolinate)copper, Bis(8-hydroxyquinolinate) manganese, tris(8-hydroxyquinolinate)aluminium, Tris (2-methyl-8-hydroxyquinolinate) aluminum, A tris (8-hydroxyquinolinate) gallium, bis(10-hydroxy benzo[h] quinolinate) beryllium, Bis(10-hydroxy benzo[h] quinolinate) zinc, a bis(2-methyl-8-quinolinate) chloro gallium, Although there are a bis(2-methyl-8-quinolinate) (o-cresolate) gallium, bis(2-methyl-8-quinolinate) (1-naphth RATO) aluminum, a bis(2-methyl-8-quinolinate) (2-naphth RATO) gallium, etc. It is not limited to these. Moreover, as a nitrogen-containing 5 member derivative, oxazole, a thiazole, oxadiazole, thiadiazole, or a triazole derivative is desirable. Specifically, it is 2 and 5-screw (1-phenyl). - 1, 3, 4-oxazole, Dimethyl POPOP, 2, 5-screw (1-phenyl) - 1, 3, 4-thiazole, 2, 5-screw (1-phenyl) - 1, 3, 4-oxadiazole, 2-(4'-tert-buthylphenyl)-5-(4"-biphenyl) 1, 3, 4-oxadiazole, 2, 5-bis(1-naphthyl)-bis[1, 3, 4-oxadiazole, 1, and 4-] [2- (5-phenyl oxadiazolyl)] benzene, 1, 4-screw [2-(5-phenyl oxadiazolyl)-4-tert-butylbenzene], 2-(4'-tert-buthylphenyl)-5-(4"-biphenyl)- 1, 3, and 4-thiadiazole -- 2,

5-bis(1-naphthyl)-bis[1, 3, 4-thiadiazole, 1, and 4-] [2- (5-phenyl thiadiazolyl)] benzene, 2-(4'-tert-buthylphenyl)-5-(4"-biphenyl)-, although there is 1, 3, 4-triazole, 2, and 5-bis(1-naphthyl)-bis[1, 3, 4-triazole, 1, and 4-] [2- (5-phenyl triazoryl)] benzene etc. It is not limited to these.

[0060] In this organic EL device, at least one sort of luminescent material, a doping ingredient, a hole-injection ingredient, and an electron injection ingredient other than the compound of a general formula [1] may contain in the same layer in a luminous layer. Moreover, it is also possible to prepare a protective layer on the surface of a component for the improvement of stability to the temperature of the organic EL device obtained by this invention, humidity, an ambient atmosphere, etc., or to protect the whole component with a silicone oil, resin, etc.

[0061] What has a bigger work function than 4eV as a conductive ingredient used for the anode plate of an organic EL device is suitable, and organic conductive resin, such as the poly thiophene and polypyrrole, is used for gold oxide groups, such as tin oxide used for those alloys, such as carbon, aluminum, vanadium, iron, cobalt, nickel, a tungsten, silver, gold, platinum, and palladium, and an ITO substrate, and a NESA substrate, and indium oxide, and a pan. Although what has a work function smaller than 4eV as conductive matter used for cathode is suitable and those alloys, such as magnesium, calcium, tin, lead,

titanium, an yttrium, a lithium, a ruthenium, manganese, and aluminum, are used, it is not limited to these. As an alloy, although magnesium/silver, magnesium/indium, a lithium/aluminum, etc. are mentioned as an example of representation, it is not limited to these. The ratio of an alloy is controlled by the temperature of the source of vacuum evaporationo, the ambient atmosphere, a degree of vacuum, etc., and is chosen as a suitable ratio. As long as an anode plate and cathode have the need, they may be formed of the lamination more than a bilayer.

[0062] In order to make light emit efficiently in an organic EL device, as for at least one side, it is desirable to make it transparence enough in the luminescence wavelength field of a component. Moreover, the transparent thing of a substrate is desirable. The above-mentioned conductive ingredient is used for a transparent electrode, and it sets it up so that predetermined translucency may secure by approaches, such as vacuum evaporationo and sputtering. As for the electrode of a luminescence side, it is desirable to make light transmittance 10% or more. Although a substrate is not limited if it has mechanical and thermal reinforcement and has transparency, it has a glass substrate and a transparency resin film. As a transparency resin film, a polyethylene and ethylene-vinyl acetate copolymerization object, An ethylene-vinyl alcohol copolymerization object, polypropylene, polystyrene, Polymethylmethacrylate, a polyvinyl chloride,

polyvinyl alcohol, A polyvinyl butyral, nylon, a polyether ether ketone, the poly ape phone, A polyether ape phon, tetrafluoroethylene-perfluoroalkyl vinyl ether, Polyvinyl fluoride, tetrafluoroethylene-ethylene, and tetrafluoroethylene-hexafluoropropylene, Polychlorotrifluoroethylene resin, poly vinylidene fluoride, polyester, a polycarbonate, polyurethane, polyimide, polyether imide, polyimide, polypropylene, etc. are raised.

[0063] Formation of each class of the organic EL device concerning this invention can apply which approach of the wet forming-membranes methods, such as the dry type forming-membranes methods, such as vacuum deposition, sputtering, plasma, and ion plating, spin coating, dipping, and flow coating. Although especially thickness is not limited, it is necessary to set it as suitable thickness. If thickness is too thick, in order to obtain a fixed optical output, big applied voltage will be needed and effectiveness will worsen. If thickness is too thin, even if a pinhole etc. will occur and it will impress electric field, sufficient luminescence brightness is not obtained. The usual thickness has the still more desirable range of 0.2 micrometers from 10nm, although the range of 10 micrometers is suitable from 5nm.

[0064] The solvent may be any, although suitable solvents, such as ethanol, chloroform, a tetrahydrofuran, and dioxane, are made to dissolve or distribute the ingredient which forms each class in the case of the wet forming-membranes

method and a thin film is formed. Moreover, also in which organic thin film layer, suitable resin and a suitable additive may be used on a membrane formation disposition for pinhole prevention of the film etc. As possible resin of use, conductive resin, such as photoconductivity resin, such as insulating resin, such as polystyrene, a polycarbonate, polyarylate, polyester, a polyamide, polyurethane, polysulfone, polymethylmethacrylate, polymethyl acrylate, and a cellulose, and those copolymers, poly-N-vinylcarbazole, and polysilane, the poly thiophene, and polypyrrole, can be mentioned. Moreover, an antioxidant, an ultraviolet ray absorbent, a plasticizer, etc. can be mentioned as an additive.

[0065] As mentioned above, organic EL device properties, such as luminous efficiency and the maximum luminescence brightness, were improvable by using the compound of this invention for the luminous layer of an organic EL device, and combining with a further specific hole injection layer or an electronic injection layer. Moreover, since it was very stable and usable luminescence brightness was obtained practical by still lower driver voltage to heat or a current, this component was also able to reduce sharply degradation which was a big problem to the former.

[0066] The organic EL device of this invention can consider application as a flat-panel display and flat-surface illuminants, such as a flat TV, to the light source of the light source of a copying machine, a printer, etc., a liquid crystal

display, instruments, etc., the plotting board, a beacon light, etc., and the industrial value is very large.

[0067] The ingredient of this invention can be used also in fields, such as an organic EL device, an electrophotography photo conductor, an optoelectric transducer, a solar battery, and image sensors.

[0068]

[Example] Hereafter, this invention is further explained to a detail based on an example.

On the glass plate with an ITO electrode washed example 1, it is compound [of Table 1] (3), 2, and 5-screw (1-naphthyl) as a luminescent material. - 1, 3, 4-oxadiazole, and polycarbonate resin (Teijin formation : panlight K-1300) were dissolved in the tetrahydrofuran by the weight ratio of 5:3:2, and the luminous layer of 100nm of thickness was obtained with the spin coating method. The electrode of 150nm of thickness was formed with the alloy which moreover mixed silver with magnesium by 10:1, and the organic EL device was obtained. As for the luminescence property of this component, luminescence of 130 (cd/m2), the highest brightness 3200 (cd/m2), and luminous efficiency 0.95 (lm/W) was obtained by direct-current-voltage 5V.

[0069] On the glass plate with an ITO electrode washed example 2, vacuum deposition of the compound (8) of Table 1 was carried out, the luminous layer of

100nm of thickness was created, the electrode of 100nm of thickness was formed with the alloy which mixed silver with magnesium by 10:1 on it, and the organic EL device was obtained. The luminous layer was vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of 10-6Torr. As for this component, luminescence of 110 (cd/m2), the highest brightness 2200 (cd/m2), and luminous efficiency 0.75 (lm/W) was obtained by direct-current-voltage 5V.

[0070] On the glass plate with an ITO electrode washed example 3, the compound (12) of Table 1 was dissolved in the methylene chloride, and the luminous layer of 50nm of thickness was obtained with the spin coating method. Subsequently, vacuum deposition of the compound (B-10) of Table 3 was carried out, the electronic injection layer of 30nm of thickness was created, the electrode of 100nm of thickness was formed on it with the alloy which mixed silver with magnesium by 10:1, and the organic EL device was obtained. The luminous layer and the electronic injection layer were vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of 10-6Torr. As for this component, green luminescence of 350 (cd/m2), the highest brightness 5400 (cd/m2), and luminous efficiency 1.3 (lm/W) was obtained by direct-current-voltage 5V.

[0071] On the glass plate with an ITO electrode washed example 4, vacuum

deposition of the compound (16) of Table 1 was carried out, and the luminous layer was formed at 50nm of thickness. Subsequently, vacuum deposition of the compound (B-3) of Table 3 was carried out, the electronic injection layer of 10nm of thickness was created, the electrode of 100nm of thickness was formed on it with the alloy which mixed silver with magnesium by 10:1, and the organic EL device was obtained. The hole injection layer and the luminous layer were vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of 10-6Torr. As for this component, green luminescence of about 410 (cd/m2), the highest brightness 10000 (cd/m2), and luminous efficiency 1.6 (lm/W) was obtained by direct-current-voltage 5V.

[0072] On the glass plate with an ITO electrode washed example 5-51, on condition that Table 4, vacuum deposition of the hole-injection ingredient was carried out, and the hole injection layer of 30nm of thickness was obtained. Subsequently, vacuum deposition of the luminescent material was carried out, and the luminous layer of 30nm of thickness was obtained. Furthermore, vacuum deposition of the electron injection ingredient was carried out, the electronic injection layer of 30nm of thickness was created, the electrode of the thickness of 150nm of thickness was formed on it with the alloy which mixed silver with magnesium by 10:1, and the organic EL device was obtained. Each class was vapor-deposited under the conditions of a substrate temperature room

temperature in the vacuum of 10-6Torr. The luminescence property of this component is shown in Table 4. Luminescence brightness here is brightness at the time of direct-current-voltage 5V seal of approval, and all the organic EL devices of this example had the high brightness property more than highest brightness 10000 (cd/m2). As a component configuration of an organic EL device, the component which combined the hole-injection ingredient of a general formula [3] and the electron injection ingredient of a general formula [4] with the luminescent material of a general formula showed the best property.

[0073]

[Table 4]

実施例	正孔注入材料	発光材料	電子注入材料	発光輝度 (cd/m²)	最大発光輝度 (cd/m²)	最大発光効率 (lm/W)
5	(A－2)	(8)	(B－3)	4420	33000	4．1
6	(A－8)	(8)	(B－3)	5100	45000	4．2
7	(A－10)	(8)	(B－3)	4800	41000	4．0
8	(A－11)	(8)	(B－3)	5200	38000	3．5
9	(A－12)	(8)	(B－3)	6000	41000	4．0
10	(A－13)	(8)	(B－3)	5800	40000	3．9
11	(A－14)	(8)	(B－3)	6000	41000	3．8
12	(A－16)	(8)	(B－3)	5500	38000	3．5
13	(A－13)	(8)	(B－11)	7700	110000	12．0
14	(A－13)	(1)	(B－11)	4300	55000	7．0
15	(A－13)	(3)	(B－11)	5000	45000	5．9
16	(A－13)	(4)	(B－11)	4400	46000	5．6
17	(A－13)	(7)	(B－11)	4600	51000	5．9
18	(A－13)	(9)	(B－11)	7100	100000	10．0
19	(A－13)	(10)	(B－11)	7600	115000	13．5
20	(A－13)	(11)	(B－11)	7200	104000	10．8
21	(A－13)	(14)	(B－11)	6800	95000	10．1
22	(A－13)	(15)	(B－11)	6500	86000	9．8
23	(A－13)	(16)	(B－11)	7100	115000	13．5
24	(A－13)	(20)	(B－11)	5800	60000	8．8
25	(A－13)	(21)	(B－11)	4900	60000	8．7
26	(A－13)	(23)	(B－11)	6800	75000	7．9
27	(A－13)	(25)	(B－11)	6500	70000	6．9
28	(A－13)	(27)	(B－11)	6800	78000	8．0
29	(A－13)	(34)	(B－11)	6500	81000	8．9
30	(A－13)	(38)	(B－11)	6900	84000	9．1
31	(A－13)	(43)	(B－11)	5800	68000	7．7
32	(A－13)	(45)	(B－11)	6100	62000	7．2
33	(A－13)	(49)	(B－11)	7500	89000	9．9
34	(A－13)	(51)	(B－11)	6000	60000	8．0
35	(A－13)	(53)	(B－11)	7100	95000	9．7
36	(A－13)	(55)	(B－11)	6900	91000	10．5
37	(A－13)	(8)	(B－1)	4500	65000	7．6
38	(A－13)	(8)	(B－4)	4100	58000	6．6
39	(A－13)	(8)	(B－9)	5000	50000	5．9
40	(A－13)	(8)	(B－12)	6900	65000	7．3
41	(A－13)	(8)	(B－13)	7500	106000	11．1
42	(A－13)	(8)	(B－15)	7000	120000	12．6
43	(A－13)	(8)	(B－16)	6200	69000	7．4
44	(A－13)	(8)	(B－18)	5500	70000	6．9
45	(A－13)	(8)	(B－20)	5900	78000	8．0
46	(A－13)	(8)	(B－21)	4300	45000	5．4
47	(A－13)	(8)	(B－22)	4000	32000	4．6
48	(A－13)	(8)	(B－23)	4500	38000	4．3
49	(A－13)	(8)	(B－24)	3300	36000	3．9
50	(A－12)	(8)	(B－14)	6000	81000	8．8
51	(A－8)	(48)	(B－18)	4100	66000	7．2

発光輝度は、素子に直流5（V）印加した時の輝度を示す。

[0074] On the glass plate with an ITO electrode washed example 52, vacuum

deposition of the hole-injection ingredient (A-13) was carried out, and the hole injection layer of 40nm of thickness was obtained. Subsequently, vacuum deposition of the compound (8) was carried out as a luminescent material, and the luminous layer of 40nm of thickness was obtained. Furthermore, vacuum deposition of (B-11) was carried out as an electron injection ingredient, and the electronic injection layer of 30nm of thickness was obtained. Moreover, the electrode of 150nm of thickness was formed for the aluminum:lithium with the alloy of the ratio of 50:1, and the organic EL device was obtained. As for this component, luminescence of 8000 (cd/m2), the highest brightness 131000 (cd/m2), and luminous efficiency 13.8 (lm/W) was obtained by direct-current-voltage 5V.

[0075] The organic EL device was produced by the same approach as an example 52 except forming the hole injection layer of 5nm of thickness of a non-metal phthalocyanine between an example 53ITO electrode and a compound (A-13). As for this component, luminescence of 10000 (cd/m2), the highest brightness 99000 (cd/m2), and luminous efficiency 10.2 (lm/W) was obtained by direct-current-voltage 5V.

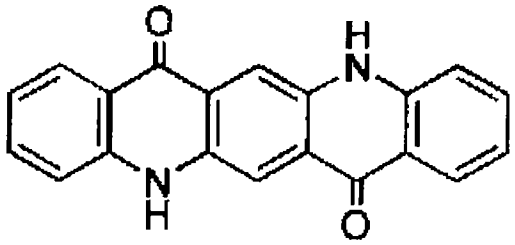
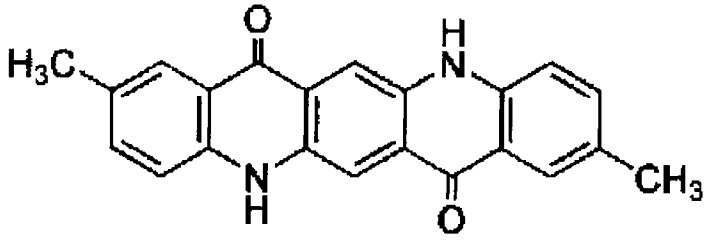
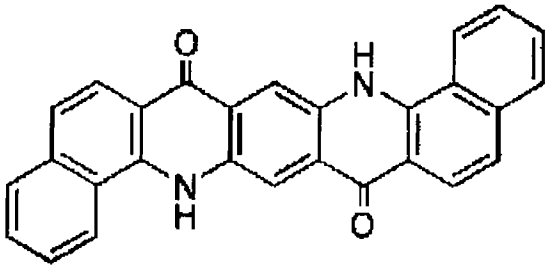
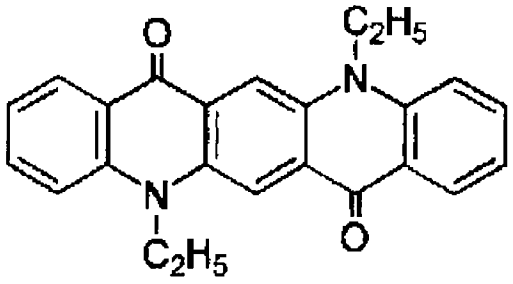
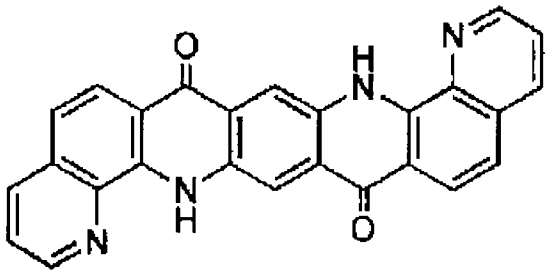
[0076] The organic EL device was produced by the same approach as an example 48 except forming the hole injection layer of 15nm of thickness of a non-metal phthalocyanine instead of example 54 compound (A-13). As for this

component, luminescence of 3500 (cd/m2), the highest brightness 77000 (cd/m2), and luminous efficiency 5.8 (lm/W) was obtained by direct-current-voltage 5V.

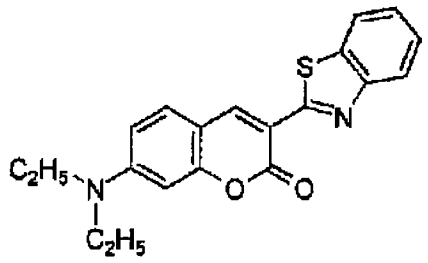
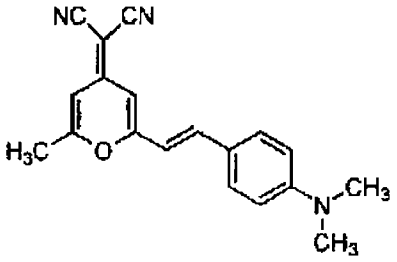
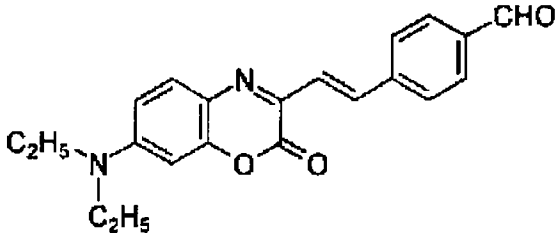
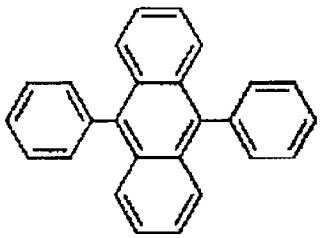
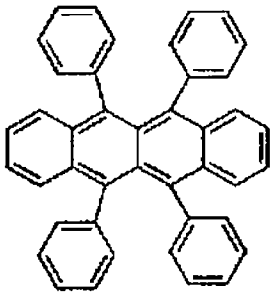
[0077] The organic EL device was produced by the same approach as an example 52 except using the luminous layer of 20nm of thickness which vapor-deposited the compound (8) and the compound shown in Table 5 by the weight ratio of 100:1 as 55 to example 64 luminous layer. The luminescence property of this component is shown in Table 6. Luminescence brightness here was brightness at the time of direct-current-voltage 5V seal of approval, and all the organic EL devices of this example have a high brightness property more than highest brightness 10000 (cd/m2), and were able to obtain the target luminescent color.

[0078]

[Table 5]

化合物	化　学　構　造
C－1	
C－2	
C－3	
C－4	
C－5	

[0079]

化合物	化　学　構　造
C－6	
C－7	
C－8	
C－9	
C－10	

[0080]

[Table 6]

実施例	表5のﾄﾞｰﾋﾟﾝｸﾞ材料	発光輝度 (cd/m^2)	最大発光輝度 (cd/m^2)	最大発光効率 (lm/W)
55	(C－1)	8300	133000	14．0
56	(C－2)	7600	105000	12．8
57	(C－3)	8500	121000	14．1
58	(C－4)	8200	100000	10．8
59	(C－5)	7700	89000	9．9
60	(C－6)	4300	51000	7．8
61	(C－7)	5500	49000	8．1
62	(C－8)	6300	55000	8．9
63	(C－9)	4800	56000	7．7
64	(C－10)	5600	78000	7．9

[0081] The organic EL device shown by this example is more than 10000 (cd/m2) as luminescence brightness, and was able to acquire high luminous efficiency altogether. About the organic EL device shown by this example, when carrying out continuation luminescence by 3 (mA/cm2), luminescence stable for 1000 hours or more could be observed, and most dark spots were not observed. Since the fluorescence quantum efficiency of luminescent material was very high, the organic EL device which used the organic EL device ingredient of this invention has improved the maximum luminescence brightness and the maximum luminous efficiency in the component which used this luminescent material by attaining high brightness luminescence in a low current seal-of-approval field, and using a doping ingredient in a luminous layer in

addition to the compound of a general formula [1]. Furthermore, the light emitting device of red luminescence or blue luminescence was able to be obtained by adding the doping ingredient of red luminescence or blue luminescence to the compound of the general formula [1] which carries out luminescence of a bluish green color, green, and yellow.

[0082] The organic EL device of this invention attains improvement in luminous efficiency and luminescence brightness, and reinforcement, and does not limit the component production approaches used collectively, such as luminescent material, a doping ingredient, a hole-injection ingredient, an electron injection ingredient, a sensitizer, resin, and an electrode material.

[0083]

[Effect of the Invention] The organic EL device which used the organic EL device ingredient of this invention as a luminescent material was able to show luminescence of the high brightness in high luminous efficiency compared with the former, and was able to obtain the long lasting organic EL device. the organic EL device formed of an organic EL device boiling further at least the compound shown by this invention by the above, and using it and the component configuration of this invention became possible [producing easily high brightness, high luminous efficiency, and a long lasting organic EL device].